EPA
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EPA-SAB-11-013 | August 2011 | www.epa.gov/sab

Reactive Nitrogen in the United States: An Analysis of Inputs, Flows, Consequences, and Management Options

A REPORT OF THE EPA SCIENCE ADVISORY BOARD

Science Advisory Board
Office of the Administrator

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UNITED STATES ENVIRONMENTAL PROTECTION AGENCY
WASHINGTON, DC 20460

August 18, 2011

EPA-SAB-11-013

OFFICE OF THE ADMINISTRATOR
SCIENCE ADVISORY BOARD

The Honorable Lisa P. Jackson
Administrator
U.S. Environmental Protection Agency
1200 Pennsylvania Avenue, N.W.
Washington, D.C. 20460

Subject: Reactive Nitrogen in the United States: An Analysis of Inputs, Flows, Consequences, and Management Options – A Report of the EPA Science Advisory Board

Dear Administrator Jackson:

Excess reactive nitrogen compounds in the environment are associated with many large-scale environmental concerns, including eutrophication of surface waters, toxic algae blooms, hypoxia, acid rain, nitrogen saturation in forests, and global warming. In addition, reactive nitrogen is associated with harmful human health effects caused by air pollution and drinking water contamination. Reactive nitrogen (hereafter referred to as Nr) includes all biologically active, chemically reactive, and radiatively active nitrogen compounds in the atmosphere and biosphere of the earth, in contrast to non-reactive gaseous N_2. EPA and other federal and state agencies have implemented programs to reduce the risks posed by excessive Nr, but a more comprehensive and integrated approach is needed to manage the use of Nr in a way to achieve its benefits, such as fertilizer for food production, and mitigate its damages as it is introduced to and cycles repeatedly through the environment in different forms and media.

The Science Advisory Board (SAB) Integrated Nitrogen Committee has conducted a study to analyze sources and fate of Nr in the United States and provide advice to EPA on integrated nitrogen research and control strategies. We are pleased to submit the SAB report, *Reactive Nitrogen in the United States: An Analysis of Inputs, Flows, Consequences, and Management Options.* Our objectives for this study were to:

- Identify and analyze from a scientific perspective the problems Nr presents in the environment and the links among them;
- Evaluate the contribution an integrated nitrogen management strategy could make to environmental protection;
- Identify additional risk management options for EPA's consideration; and
- Make recommendations to EPA concerning improvements in nitrogen research to support risk reduction.

The SAB report provides findings and recommendations addressing these study objectives. Assessment of the challenges and costs to the Agency of implementing the recommendations is beyond the scope of the report.

In general, the SAB finds that:

- In the United States, human activities across multiple sources currently introduce more than five times the Nr into the environment than natural processes. The largest U.S. sources of new Nr entering the U.S. environment include: the creation and use of synthetic fertilizers, Nr created by legumes, and the combustion of fossil fuels.
- Much of the Nr used to ensure a plentiful supply of food, fiber and biofuel is released to the environment, as is the Nr formed during fossil fuel combustion.
- The introduction of human created Nr into the environment degrades air and water quality, which can cause harmful algae blooms, hypoxia, fish kills, loss of drinking water potability, loss of biodiversity, forest declines, and human health problems resulting in losses of billions of dollars per year.

- Multiple strategies and actions exist to more effectively minimize the inputs of Nr to the environment and maximize nitrogen use efficiency.

The SAB provides the following overarching recommendations to improve the management of Nr.

- The framing of the movement of nitrogen among various environmental reservoirs in terms of the nitrogen cascade concept provides a means for tracking nitrogen as it changes form and passes through multiple ecosystems and media. Given this complexity, innovative management systems and regulatory structures reflecting these characteristics of Nr are required to address the significant environmental and human health damage caused by Nr. New institutional structures and relationships that also reflect the multi-media and multi-form character of Nr and its flows and transformations through the environment will have to be created for effective control and management.
- The SAB recommends an integrated approach to the management of Nr. This approach must use a combination of implementation mechanisms appropriate to the specific environmental and policy contexts and supported by critical research on the specific risks of Nr and on decreasing the risks of Nr. The approach must reflect an integrated policy that recognizes the complexity and trade-offs associated with the nitrogen cascade while recognizing that intervention points vary in terms of efficiency and cost effectiveness.
- EPA should form an intra-Agency Nr management task force that will build on the existing breadth of Nr research and management capabilities within the Agency. Its objective should be to increase scientific understanding of: (1) Nr impacts on terrestrial and aquatic ecosystems, human health, and climate; (2) Nr-relevant monitoring requirements; and (3) the most efficient and cost effective means by which to decrease various adverse impacts of Nr loads as they cascade through the environment.
- Successful Nr management will require changes in the way EPA interacts with other agencies. The SAB recommends that EPA convene a reactive nitrogen inter-agency management task force with broad representation from other agencies and departments involved with Nr control or utilization. This is essential to coordinate federal programs that address Nr concerns and would help ensure clear responsibilities for monitoring, modeling, researching, and managing Nr in the environment. Similar efforts at coordination and joint action need to be made among and between agencies at both the state and federal level.

In the context of addressing the specific study objectives, the SAB explored how an estimated 25 percent reduction in Nr introduced into the environment might be achieved with existing technology in the coming 10 to 20 years through actions that could be taken by EPA, other management authorities, and other public and private organizations. Specific actions include increased controls of oxides of nitrogen, improved reactive nitrogen uptake by agricultural crops, large-scale creation and restoration of wetlands for nitrogen removal in agricultural landscapes with high Nr in surface waters, decreased loss of reactive nitrogen from agricultural lands and animal feeding operations, and decreased discharge of reactive nitrogen from point sources and developed (urban) lands. However, dealing effectively with reactive nitrogen's cascade through air, water and land will require an integrated management approach that is multi-media and multi-stressor as suggested by recent initiatives by EPA's Office of Research and Development.

The most important task for EPA and allied agencies and departments will be to effectively inform the public of the costs and dangers of excess Nr. Without strong public support, the widespread efforts necessary to control Nr will not be possible.

In closing, we appreciate the opportunity to provide advice on this very important topic, and we look forward to receiving your response. The SAB stands ready to provide more information as it may be useful and would be pleased to assist EPA in the implementation of the report's recommendations, if the EPA would find such support valuable.

Sincerely,

/Signed/
Dr. Otto C. Doering III
Chair
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Science Advisory Board

/Signed/
Dr. James N. Galloway
Chair (2007-2009)
Integrated Nitrogen Committee
Science Advisory Board

/Signed/
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Vice-Chair (2008-2009)
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Science Advisory Board

/Signed/
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Dr. Paige Tolbert, Professor and Chair, Department of Environmental Health, Rollins School of Public Health, Emory University, Atlanta, GA

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Dr. Robert Watts, Professor of Mechanical Engineering Emeritus, Tulane University, Annapolis, MD

Dr. R. Thomas Zoeller, Professor, Department of Biology, University of Massachusetts, Amherst, MA

SCIENCE ADVISORY BOARD STAFF

Dr. Angela Nugent, Designated Federal Officer, U.S. Environmental Protection Agency, Washington, DC

Acknowledgements

The EPA Science Advisory Board Integrated Nitrogen Committee would like to acknowledge many individuals who provided their perspectives and insights for the Committee's consideration in the development of this report.

Invited speakers provided perspectives at public meetings of the Committee. These speakers included: the following individuals from the U.S. EPA - Mr. Robert Bastian, Mr. Gilbert Castellanos, Mr. John Davies, Dr. Robin Dennis, Dr. Jonathan Garber, Dr. Richard Haeuber, Dr. Alan Hecht, Mr. Rudolph Kapichak, Mr. Gary Lear, Dr. Richard Linthurst, Dr. Rohit Mathur, Ms. Roberta Parry, and Mr. James Pendergast; the following individuals from the USDA - Dr. Roger Claassen, Dr. Stan Daberkow, Dr. Raymond Knighton, and Dr. Mark Walbridge; and the following individuals from other organizations - Mr. Craig Cox, Soil and Water Conservation Society; Dr. Jan Willem Erisman, Energy Research Centre of the Netherlands; Dr. Paul Fixen, International Plant Nutrition Institute; Dr. David McNaught, Environmental Defense Fund; Dr. John Miranowski, Iowa State University; Dr. Martin Petrovic, Cornell University; and Mr. John Sheehan, LiveFuels.

The Committee would also like to thank the speakers and participants who attended the SAB Integrated Nitrogen Committee workshop held on October 20-22, 2008. These participants included: the following individuals from the U.S. EPA - Mr. Richard Batiuk, Dr. Jana Compton, Dr. Robin Dennis, Ms. Katie Flahive, Dr. Richard Haeuber, Ms. Chris Lewiciki, Dr. James Liebman, Ms. Sally Shaver, Ms. Roberta Parry, Mr. Randy Waite, Mr. John Wilson, Mr. Tom Wirth, and Mr. Marcus Zobrist; the following individuals from the USDA - Dr. Dan Jaynes, Dr. Ray Knighton, and Mr. Richard Swenson; and the following individuals from other organizations - Dr. Jan Willem Erisman, Energy Research Centre of the Netherlands; Mr. Albert Ettinger, Environmental Law and Policy Center; Mr. John Hardin, John Hardin & Son; Dr. Shelie Miller, University of Michigan; Dr. Catherine O'Connor, Metropolitan Water Reclamation District of Greater Chicago; Mr. Robin O'Malley, The Heinz Center; Mr. Richard Poirot, Vermont Agency of Natural Resources; Mr. John Quinn, Constellation Energy; Mr. Pat Rice, Nebraska Department of Environmental Quality; Dr. Joe Rudek, Environmental Defense Fund; Dr. Ted Russell, Georgia Institute of Technology; Ms. Mindy Selman, World Resources institute; Dr. Thomas Simpson, University of Maryland; Dr. Richard Smith, U.S. Geological Survey; Dr. Clifford Snyder, International Plant Nutrition Institute; Dr. Robert Summers, Maryland Department of the Environment; Dr. David Whitall, National Oceanic and Atmospheric Administration; Ms. Marcia Willhite, Illinois Environmental Protection Agency; and Mr. James Wurtz, Railway Equipment Company.

In addition, the Committee thanks the experts who provided independent review of the report in draft form: Dr. John Day, Louisiana State University; Dr. Elisabeth Holland, National Center for Atmospheric Research; Dr. Gregory McIsaac, University of Illinois; Dr. Jerry Melillo, Marine Biological Laboratory, Woods Hole, Massachusetts; Dr. Gyles Randall, University of Minnesota; Dr. James Schauer, University of Wisconsin-Madison; and Dr. Stuart Weiss, Creekside Center for Earth Observations. The reviewers provided many constructive comments and suggestions, but were not, however, asked to endorse the conclusions or recommendations in the report.

NOTICE

This report has been written as part of the activities of the EPA Science Advisory Board (SAB), a public advisory group providing extramural scientific information and advice to the Administrator and other officials of the Environmental Protection Agency. The SAB is structured to provide balanced, expert assessment of scientific matters related to problems facing the Agency. This report has not been reviewed for approval by the Agency and, hence, the contents of this report do not necessarily represent the views and policies of the Environmental Protection Agency, nor of other agencies in the Executive Branch of the Federal government, nor does mention of trade names of commercial products constitute a recommendation for use. Reports of the SAB are posted on the EPA website at http://www.epa.gov/sab.

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List of Chemical Abbreviations

C	Carbon
CFC	Chlorofluorocarbon
DIN	Dissolved inorganic nitrogen
DO	Dissolved oxygen
Fe	Iron
H	Hydrogen
HNO_3	Nitric acid
HONO	Nitrous acid
N	Nitrogen
N_2	Diatomic (molecular) nitrogen
N_2O	Nitrous oxide,
N_2O_5	Dinitrogen pentoxide (nitric acid anhydride)
NH_3	Ammonia
NH_4^+	Ammonium
NH_x	NH_3 NH_4^+
NO	Nitric oxide
NO_2	Nitrogen dioxide
NO_3^-	Nitrate ion
NO_3	Nitrate radical
N_{org}	Organic nitrogen
NO_x	Nitrogen oxides (NO + NO_2)
NO_y	Total reactive oxidized nitrogen (NO, NO_2, NO_3, 2xN_2O_5, HONO, HNO_3, NO_3^-, PAN and other organo-nitrates, $RONO_2$)
Nr	Reactive nitrogen
O_2	Oxygen
OH	Hydroxyl radical
P	Phosphorus
PAN	Peroxy acetyl nitrate
PM	Particulate matter
$PM_{2.5}$	Particulate matter less than 2.5 microns in diameter
PM_{10}	Particulate matter less than 10 microns in diameter
$RONO_2$	Organic nitrates
Si	Silicon
SO_2	Sulfur dioxide
SO_4^{2-}	Sulfate
TAN	Total ammonical nitrogen

List of Acronyms and Abbreviations

AAPFCO	Association of American Plant Food Control Officials
AARA	American Reinvestment and Recovery Act
AIRMON	Atmospheric and Integrated Research Monitoring Network
AOB	Ammonia oxidizing bacteria
BL	Boundary layer
BMP	Best management practice
BNF	Biological nitrogen fixation
BNR	Biological nutrient (or nitrogen) removal
CAA	Clean Air Act
CAFO	Concentrated animal feeding operation
CAIR	Clean Air Interstate Rule
CALM	Consolidated Assessment and Listing Methodology
CAST	Council for Agricultural Science and Technology
CASTNET	Clean Air Standards and Trends Network
C-BNF	Cultivation-induced biological nitrogen fixation
CCC	Criterion continuous concentration
CFC	Chlorofluorocarbon
CFR	Code of Federal Regulations
CL	Critical load (threshold of Nr loading at which negative impacts have been documented)
CLAD	Critical Loads Ad-Hoc Committee
CMAQ	Community multiscale air quality
CMC	Criterion maximum concentration
CRP	Conservation Reserve Program
CSO	Combined sewer overflow
CTM	Chemical Transport Models
CWA	Clean Water Act
CWSRF	Clean Water State Revolving Fund (construction grants program under the Clean Water Act)
DOE	U.S. Department of Energy
DOT	U.S. Department of Transportation
EGU	Electricity generating units
EFD	Essential Facilities Doctrine
EGR	Exhaust gas recirculation
EISA	Energy Independence and Security Act
EPA	United States Environmental Protection Agency
EQIP	Environmental Quality Incentives Program
EU	European Union
FAO	Food and Agricultural Organization of the United Nations
FAOSTAT	Food and Agricultural Organization Statistical Database
FGR	Flue-gas recirculation

ha	Hectare
GHG	Greenhouse gas
GPS	Geographic Positioning System
HAB	Harmful algal bloom
IPCC	Intergovernmental Panel on Climate Change
ISA	Integrated Science Assessments
ITQ	Individual transferable quota
kg	Kilogram
L	Liter
LA	Load allocation
LCA	Life cycle analysis
LISS	Long Island Sound Study
MCL	Maximum contaminant level
MCLG	Maximum contaminant level goal
mg	Milligrams
MGD	Million gallons per day
MJ	Megajoule (one million joules)
Mmt	Million metric tons
MT	metric tons
MOM	Mississippi-Ohio-Missouri
MRB	Mississippi River Basin
MS4	Municipal separate storm sewer system
NAAQS	National Ambient Air Quality Standards
NADP	National Atmospheric Deposition Program
NASS	National Agricultural Statistics Service Information
NCA	National Coastal Assessment
NCE	Nitrogen Credit Exchange
NCCR	National Coastal Condition Report
NEEA	National Estuarine Eutrophication Assessment
NESCAUM	Northeast States for Coordinated Air Use Management
NFUE	Nitrogen fertilizer use efficiency. Calculated as the ratio of grain yield to the quantity of applied N fertilizer (kg grain/kg applied N).
NMP	Nutrient management plan
NOAA	National Oceanic and Atmospheric Administration
NPS	Nonpoint source
NRC	National Research Council
NRCS	Natural Resources Conservation Service
NRD	Natural Resource District
NRI	National Resources Inventory
NTN	National Trends Network

NUE	Nitrogen use efficiency. NUE is defined as the kg grain produced per kg of total N used by the crop, where total N includes N from fertilizer, biological N fixation, and soil organic matter mineralization
OTAG	Ozone Transport Assessment Group
OTC	Ozone Transport Commission
PE	Physiological efficiency (physiological efficiency with which the N taken up by the crop is used to produce economic yield such as grain or fruit, quantified by kg increase in economic yield per kg of N accumulation in above ground crop biomass)
PFP	Partial factor productivity
POTW	Publicly owned treatment works
PSD	Prevention of significant deterioration
RE	Recovery efficiency (kg N uptake per kg N applied)
SAV	Submerged aquatic vegetation
SNCR	Selective non-catalytic reduction
SCR	Selective catalytic reduction
SIP	State Implementation Plan
SOM	Soil organic matter
SPARROW	Spatially Referenced Regressions on Watershed Attributes Model
STP	Sewage treatment plant
SW	Storm water
SWAT	Storm Water Assessment Tool
SWPPP	Stormwater Pollution Prevention Plan
T	Temperature
Tg	Teragram (million metric tons or 10^{12} grams)
TMDL	Total maximum daily load
TN	Total nitrogen
UFTRS	Uniform Fertilizer Tonnage Reporting System
UNECE	United Nations Economic Commission for Europe
USDA	U.S. Department of Agriculture
USGS	U.S. Geological Survey
USEPA	United States Environmental Protection Agency
WHO	World Health Organization
WLA	Wasteload allocation
WPCA	Water pollution control authorities
WRI	World Resources Institute
WRP	Wetland Reserve Program
WSA	Wadeable Stream Assessment

Executive Summary

Introduction

Nitrogen (N) is an integral component of all proteins, which are the basic building blocks of life and catalysts for life-sustaining reactions in organisms. Reactive nitrogen (Nr), in contrast to non-reactive gaseous N_2, includes all biologically active, chemically reactive, and radiatively active nitrogen compounds in the atmosphere and biosphere of the earth.[1] Without an adequate supply of N in any organism's diet, it can't survive. Ironically, bioavailable N for nutrition is in short supply and indeed the productivity of most of the world's ecosystems is often limited by the availability of N. This is certainly the situation with food production. Without the creation of N fertilizer by an industrial process (the Haber-Bosch process) and the increased cultivation of leguminous crops, the world could not support the current human population or its projected increase.

As further discussed in Chapter 2 of this report, increased anthropogenic input of Nr to the environment has contributed to large increases in the mass flux of nitrogen via the nitrogen cycle. Anthropogenic sources of N now provide enough N, on average, to grow food for the world's peoples. However, a major consequence of this nearly inexhaustible supply is that most N used in food production, and all of the new Nr produced by fossil fuel combustion, is lost to the environment where it circulates through the earth's atmosphere, hydrosphere, geosphere, and biosphere. During this circulation, Nr contributes to a wide variety of consequences, which are magnified with time as Nr moves through the environment.

Impacts of reactive nitrogen on human health and the environment

Anthropogenic creation of Nr provides essential benefits for humans – first and foremost in meeting human dietary needs. A large fraction of the human population of the earth could not be sustained if synthetic nitrogen fertilizers did not significantly augment food production. Essentially all of the Nr created by human activities, however, is released to the environment, often with unintended negative consequences. As summarized in Table ES-1, it contributes to a number of adverse public health and environmental effects, including photochemical smog, decreased atmospheric visibility, acidification of terrestrial and aquatic ecosystems, eutrophication of coastal waters (i.e., harmful algal blooms, hypoxia), drinking water concerns, freshwater Nr imbalances, greenhouse gas emissions and subsequent climate change, and stratospheric ozone depletion.

In light of the magnitude of the human alteration of the nitrogen cycle, and the resulting negative consequences on humans and ecosystems, the National Academy of Engineering has identified management of the nitrogen as one of the "grand challenges" facing this country.[2]

Nr effects are manifest as direct declines in both human health (e.g., respiratory and cardiac diseases) and ecosystem health (e.g., coastal eutrophication and loss in biodiversity). In addition, there are indirect declines in human health because the negative impacts on ecosystems will diminish the services that those ecosystems provide people. The effects are often magnified because the same atom of nitrogen can cause multiple effects in the atmosphere, in terrestrial ecosystems, in freshwater and marine systems, and on human health. We call this sequence of effects the nitrogen cascade.

The nitrogen cascade

The nitrogen cascade has three dimensions: biogeochemical, alterations in the environment, and human and ecosystem consequences.

The "biogeochemical" dimension of the nitrogen cascade involves: Nr creation from N_2 as a consequence of chemical, food, and energy production; Nr use in food and chemical production; Nr losses to the environment; changes in Nr species residence times in environmental reservoirs; Nr transfers among reservoirs; and Nr conversion back to N_2. Alterations to the environment then result from increased Nr levels in the environment. These alterations have negative consequences for ecosystem and human health at local, regional, national, and global scales. Because nitrogen is a critical resource and also a contributor to many of the environmental concerns facing the U.S. today, it is imperative to understand how human action has altered N cycling in the U.S., and the consequences of those alterations on people and ecosystems. The overarching question is, how do we protect and sustain ecosystems that provide multiple benefits to society while also providing the interconnected material, food and energy required by society?

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[1] Reactive nitrogen (Nr) includes inorganic chemically reduced forms of N (NH_x) [e.g., ammonia (NH_3) and ammonium ion (NH_4^+)], inorganic chemically oxidized forms of N [e.g., nitrogen oxides (NO_x), nitric acid (HNO_3), nitrous oxide (N_2O), N_2O_5, HONO, peroxy acetyl compounds such as peroxyacytyl nitrate (PAN), and nitrate ion (NO_3^-)], as well as organic compounds (e.g., urea, amines, amino acids, and proteins).

[2] National Academy of Engineering Grand Challenges (http://www.engineeringchallenges.org/cms/challenges.aspx)

Table ES-1: Examples of impacts of excess reactive nitrogen on human health and environment

Impact	Cause	Location	Metric	Source	Reference
AIR					
Visibility decrease	Fine particulate matter	National Parks and wilderness areas	visibility impairment	NO_y and NH_x from fossil fuels and agriculture	Malm et al., 2004; U.S. EPA Clean Air Scientific Advisory Committee, 2004 EPA-CASAC-09-010
LAND - ECOSYSTEMS					
Biodiversity loss	Nitrogen deposition	Grasslands and forests in the United States receiving N deposition in excess of critical load	Decrease in species richness of grasslands and forests	Utilities, traffic, and animal agriculture	Bobbink et al., 2010; Fenn et al., 2003.
Forest decline	Ozone and acid deposition	Eastern and Western United States	Decreased timber growth; increased susceptibility to disease and pests	Utilities, traffic, and animal agriculture	Johnson & Siccama, 1983; MacKenzie & El-Ashry, 1990
LAND - AGRICULTURE					
Crop yield loss	Ozone	Eastern and Western United States	$ 2-5 billion/year	Utilities & traffic	Heck et al., 1984
WATER					
Acidification of surface waters; loss of biodiversity	Acidification of soils, streams and lakes is caused by atmospheric deposition of sulfur, HNO_3, NH_3 and ammonium compounds.	Primarily mountainous regions of the United States	Out of 1,000 lakes and thousands of miles of streams in the Eastern United States surveyed, 75% of the lakes and 50% of the streams were acidified by acid deposition	Fossil fuel combustion and agriculture	U.S. EPA, 2008a http://www.epa.gov/acidrain
Hypoxia of coastal waters	Excess nutrient loading, eutrophication, variable freshwater runoff	Gulf of Mexico, other estuarine and coastal waters	Benthic finfish/shellfish habitat loss, fish kills, sulfide toxicity, costs >$50 million annually	N, P from energy and food production	Bricker et al., 1999; Verity et al., 2006; U.S. EPA SAB, 2007; Rabalais et al., 1999; Mitsch et al., 2001
Harmful Algal Blooms	Excessive nutrient loading, climatic variability	Inland and coastal waters	Fish kills, losses of drinking and recreational waters costs >$100 million annually	Excess nutrient (N & P) loading	Paerl, 1988; ECOHAB, 1995; NRC, 2000
HUMAN AND ENVIRONMENTAL HEALTH DAMAGES					
Human mortality	$PM_{2.5}$, O_3 and related toxins.	U.S. urban and nearby areas	Pollution related deaths estimated at 28,000-55,000 per year (a range of cardiovascular and respiratory system effects are associated with this pollution).	NO_y and NH_x from fossil fuels and agriculture	Mokdad et al., 2004; Ezzati et al., 2004.
Total damage to public health and environment	NO_x into air	Chesapeake Bay Watershed	$3.4 Billion; 200,000 MT	Mobile sources	Moomaw and Birch, 2005; Birch et al., 2011
Total damage to public health and environment	NH_x and nitrate into air and water	Chesapeake Bay Watershed	$1.5 Billion; 400,000 MT	Agriculture	Moomaw and Birch, 2005

Nr inputs to the nation and the world have been increasing, largely due to human activities associated with food production and fossil fuel combustion. Despite the obvious benefits of a plentiful supply of food and energy, the adverse consequences associated with the accumulation of Nr in the environment are large, with implications for human health and the environment.

The greater the inputs of Nr to the landscape, the greater the potential for negative effects caused by greenhouse gas (GHG) production, ground level ozone, acid deposition, and Nr overload that can contribute to climate change, degradation of soils and vegetation, acidification of streams, lakes and rivers, estuarine and coastal eutrophication, hypoxia, and habitat loss.

The growing nature of the Nr problem, and the adverse and intertwined consequences associated with Nr inputs to air, land, and water as exhibited in the N cascade underscore the need for researchers and managers to explore integrated strategies that minimize N inputs, maximize its use efficiency, promote Nr removal processes, and protect humans and natural resources.

The concept of the nitrogen cascade highlights that once a new Nr molecule is created, it can, in sequence, travel throughout the environment contributing to major environmental problems (Galloway et al., 2003). The adaptation of the cascade in Figure ES-1 was developed by the SAB Integrated Nitrogen Committee (INC) to provide a context for considering nitrogen-related issues and ecosystem effects in the U.S. To consider the cascading effects of Nr in the U.S., we examined the various atmospheric, terrestrial, and aquatic environmental systems where Nr is stored, and the magnitudes of the various flows of N to, from, and within them. The nitrogen cascade concept implies the cycling of Nr among these systems. The process of denitrification is the only mechanism by which Nr is converted to chemically inert N_2, "closing" the continuous cycle (Figure ES-1 shows only flows of reactive nitrogen, not N_2). Denitrification can occur in any of the indicated reservoirs except the atmosphere.

The "new" N box in the Nitrogen Cascade depicts the two primary anthropogenic sources by which Nr originates – energy production and food production – and where Nr from these sources enters ecosystems. Energy production includes both fossil fuel and biofuel combustion. Food production includes N fertilizer produced in the U.S., cultivation-induced biological N (C-BNF) in the U.S., production of animals and crops in the U.S. for human consumption, and imports of N-containing fertilizer, grain and meat to the U.S.

The Nitrogen Cascade

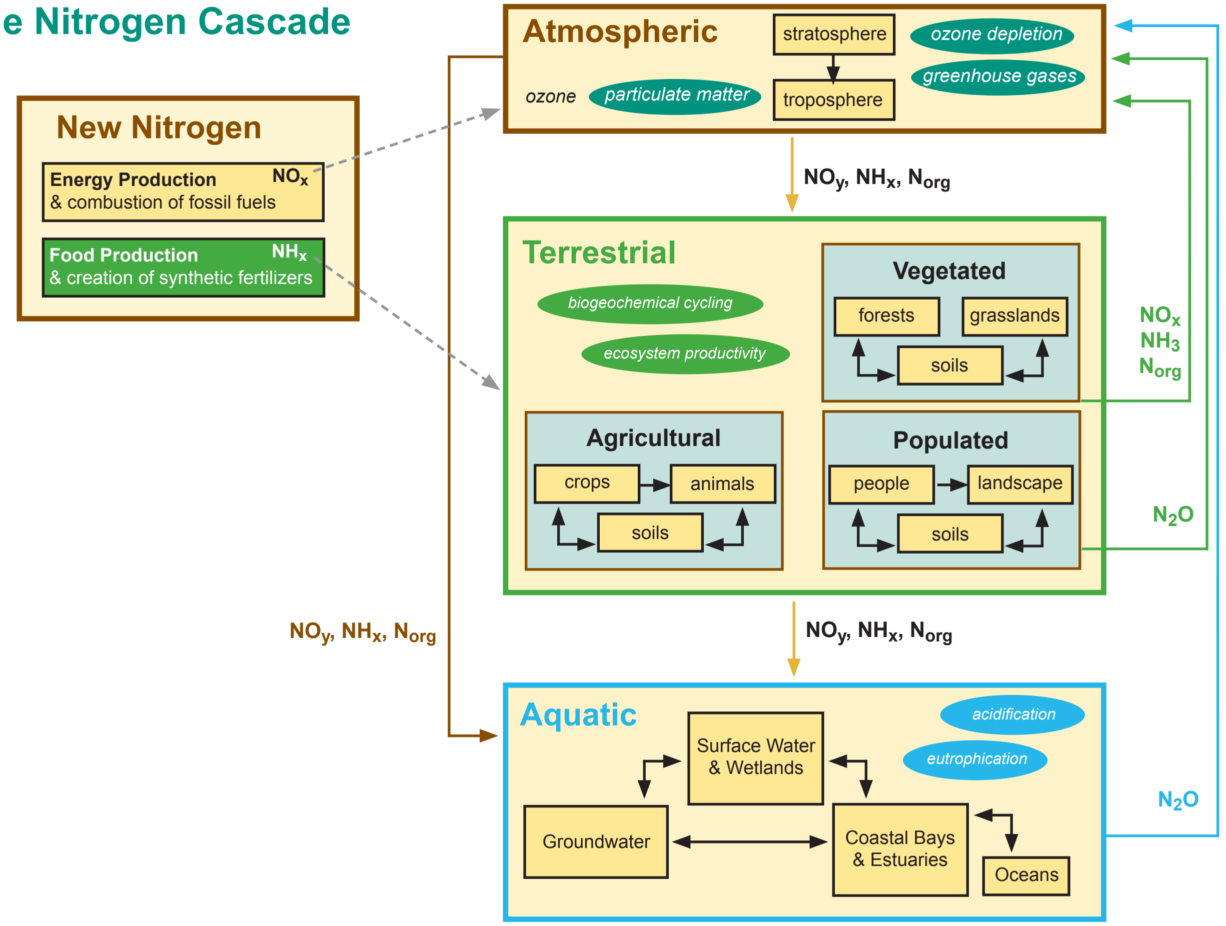

Figure ES-1: The nitrogen cascade

The atmospheric system box in the Figure ES-1 indicates that tropospheric concentrations of both ozone and particulate matter are increased due to emissions of nitrogen oxides[3] (NO_x) to the atmosphere. The ovals illustrate that the increase in N_2O concentrations, in turn, contribute to the greenhouse effect in the troposphere and to ozone depletion in the stratosphere. Except for N_2O, there is limited Nr storage in the atmosphere. Losses of Nr from the atmospheric system include total oxidized nitrogen[4] (NO_y), reduced nitrogen[5] (NH_x), and organic nitrogen (Norg) deposition to terrestrial and aquatic ecosystems of the earth's surface. There is little potential for conversion of Nr to N_2 via denitrification in air. However, once airborne deposition of Nr occurs it will be subject to denitrification pathways via soil and water.

The terrestrial system box in the Figure ES-1 depicts that Nr enters agricultural lands via food production and is introduced to the entire terrestrial landscape via atmospheric deposition. Within agricultural regions there is cycling among soils, crops and animals, and then a transfer of Nr as food to populated regions, from which there are Nr losses to the environment (e.g., sewage, landfills). The ovals showing ecosystem productivity and biogeochemical cycling reflect that Nr is actively transported and transformed within the terrestrial system, and that as a consequence there are significant impacts on ecosystem productivity due to fertilization and acidification, often with resulting losses of biodiversity. There is ample opportunity for Nr storage in both biomass and soils. Losses of Nr from this system occur by leaching and runoff of NO_y, NH_x and Norg to aquatic ecosystems and by emissions to the atmospheric system as NO_x, NH_3, Norg, and N_2O. There is potential for conversion of Nr to N_2 via denitrification in the terrestrial system.

The aquatic system box in the Figure ES-1 shows that Nr is introduced via leaching and runoff from terrestrial ecosystems and via deposition from atmospheric ecosystems. Connected with the hydrological cycle, there are Nr fluxes downstream with ultimate transport to coastal systems. Within the aquatic system, the ovals highlight two significant impacts of waterborne Nr acidification of freshwaters and eutrophication of fresh and coastal waters. Except for Nr accumulation in groundwater reservoirs, there is limited Nr storage within the hydrosphere. Losses of Nr from the aquatic system are primarily via N_2O emissions to the atmospheric system. There is a very large potential for conversion of Nr to N_2 via denitrification in water and wetlands.

NO_y, NH_x and N_2O are all components of Nr, but a fundamental difference is that the NO_y and NHx are rapidly transferred from the atmosphere to receiving ecosystems due to a short atmospheric residence time (≤ 10 days) where they continue to contribute to the N cascade. Because of its longer residence time (~100 years) however, N_2O remains in the troposphere where it contributes to climate change, until it is transferred to the stratosphere, where it contributes to ozone depletion.

Trends in N inputs to the United States

In 2002, humans introduced 29 teragrams (Tg) of newly formed reactive N into the U.S. through Haber-Bosch process production of fertilizers and industrial Nr, cultivation-induced biological nitrogen fixation (i.e., conversion of N_2 to NH_3 by microorganisms associated with some cultivated crops, for example, legumes), and fossil fuel combustion (Figure ES-2). By definition, prior to human presence in the U.S., there was no introduced anthropogenic Nr. Prior to 1900, no Haber-Bosch Nr was introduced, fossil fuel combustion introduced very small amounts relative to today, and cultivation-induced biological nitrogen fixation created approximately 2 Tg N. Thus, between 1900 and 2002, the amount of Nr introduced to the U.S. has increased by approximately 10-fold.

Nitrogen inputs to the United States

The EPA Science Advisory Board (SAB) Integrated Nitrogen Committee ("Committee") evaluated nitrogen inputs to the U.S. in 2002. At the global scale, human activities produced approximately twice as much Nr as did natural processes. In the U.S., however, the amount of Nr produced by human activities was approximately five times larger than natural processes. As shown in Figure ES-2, natural ecosystems in the U.S. introduce about 6.4 Tg of Nr as N per year (Tg N/yr). In contrast, human activities introduce about 28.5 Tg N/yr.

Chapter 2 of this report discusses sources, transfer, and transformation of Nr. Supporting references for the information presented on this topic are presented in Chapter 2. The largest single source of Nr in the U.S. is the Haber-Bosch process, which introduces about 15.2 Tg N/yr: 9.4 Tg N/yr from domestic Nr production and 5.8 Tg N/yr from imports of Nr in fertilizers. The 15.2 Tg N/yr of anthropogenic Nr is used in three ways: 9.9 Tg N/yr is used to produce agricultural crops; 1.1 Tg N/yr is applied to turf grasses; and 4.2 Tg N/yr is used by industry for production of nylon, refrigerants, explosives and other commercial products.

The second largest source of Nr introduced into the U.S. is enhancement of biological nitrogen fixation (BNF) by cultivation of legumes like soybeans and alfalfa that have nitrogen-fixing symbionts, or by crops like rice that have nitrogen-fixing bacteria in their rhizosphere. These Nr fixing crops introduce about 7.7 Tg N/yr. A

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[3] NOx (oxides of nitrogen) includes NO + NO_2

[4] NOy (total oxidized nitrogen) includes NO, NO_2, NO_3, N_2O_5, HONO, HNO_3, NO_3–, PAN and other organo-nitrates, $RONO_2$

[5] NHx (reduced nitrogen) includes NH_3 + NH_4

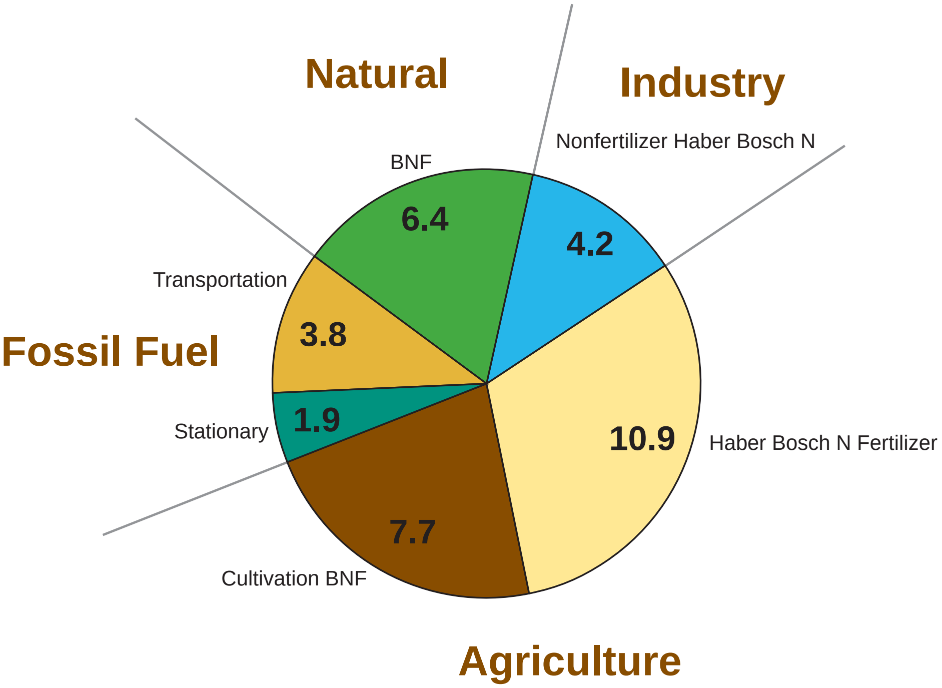

Figure ES-2: Sources of reactive nitrogen (Nr) introduced into the United States in 2002 (Tg N/yr).

Figure ES-2 explanatory notes:

Numerical units: teragram of reactive nitrogen (Nr) per year (Tg N/yr)

Natural BNF: biological nitrogen fixation in natural grasslands, rangelands, and forests,

Fossil Fuel-Transportation: combustion in vehicles, trains, airplanes, ships and off-road construction equipment.

Fossil Fuel-Stationary: combustion of fossil fuels in power plants and industrial boilers.

Agriculture-cultivation BNF: agricultural augmentation of biological nitrogen fixation – for example by planting of nitrogen fixing legumes.

Agriculture-Haber Bosch N fertilizer: agricultural (including turf production) use of synthetic nitrogen fertilizers produced by the Haber Bosch process for converting gaseous N_2 to Nr.

Industry-Haber Bosch N: Industrial sources of Nr produced by the Haber-Bosch process.

Figure ES-2 documents only the introduction of new Nr in the United States, and not the transfers of existing Nr among systems (e.g., Nr in manure).

small amount of additional Nr is also imported in grain and meat products; in 2002 this source of added Nr was approximately 0.2 Tg N/yr (not shown in Figure ES-2).

Fossil fuel combustion is the third largest source of new Nr. It introduces approximately 5.7 Tg N/yr into the environment (almost entirely as NO_x), that is, 3.8 Tg N/yr from transportation sources and 1.9 Tg N/yr from stationary sources such as electric utilities, industrial boilers and from certain industrial processes.

In summary, agriculture and domestic use of fertilizers to produce food, feed, and fiber (including bioenergy and BNF) and combustion of fossil fuels are the largest sources of new Nr released into the environment in the U.S. The percentage distribution of Nr released to the U.S. environment from human activities in 2002 was: about 65% from agricultural sources (including BNF and turf production), about 20% from fossil fuel sources, and about 15% from industrial sources (Figure ES-2).

Distribution of reactive nitrogen through the environment

Once introduced into the U.S., Nr compounds are distributed via the atmosphere, hydrosphere, geosphere, biosphere, and commerce. Distribution in the atmosphere begins with NO_x, NH_3, and N_2O. NO_x and NH_3 (and their reaction products) are distributed on a scale of hundreds to thousands of kilometers within the U.S. boundaries, and also distributed to downwind countries and oceans. Due to its long lifetime (approximately 100 years) in the atmosphere, N_2O accumulates in the U.S. atmosphere and is also dispersed throughout the global atmosphere. All ecosystems in the conterminous U.S. receive anthropogenic Nr from the atmosphere and for many ecosystems it is their primary, albeit unintended, source of Nr. Once deposited, Nr can be stored in soils and biomass and widely distributed via the stream-river continuum to inland and coastal waters. Some of the Nr is converted to N_2O or denitrified to N_2, primarily in aquatic ecosystems, including wetlands. Commerce is a major mechanism that transfers Nr from one place to another in the U.S.; most of the Nr that is used to produce food (e.g., fertilizer) and in food products crosses state boundaries via roads, railroads and the air.

Putting values to this distribution, of the 6.3 Tg N/yr of U.S. NO_x emissions, 2.7 Tg N/yr are deposited back onto the land and surface waters of the U.S. Thus, by difference we estimate that as much as 3.6 Tg N/yr of the U.S. NO_x emissions are advected out of the U.S. via the atmosphere. Similarly, of the 3.1 Tg N/yr of NH_3 that are emitted into the U.S. atmosphere each year, about 2.1 Tg N/yr are deposited onto the land and surface waters of the U.S., and about 1 Tg N/yr is advected out of the U.S. via the atmosphere. Emissions of N_2O discharge about 0.8 Tg N/yr into the global atmosphere. In sum, 5.4 Tg N are advected out of the U.S. from all sources each year either to other nations or to the global atmospheric or ocean commons.

Riverine discharges of Nr to the U.S. coastal zone account for 4.8 Tg N/yr, while export of N-containing commodities (e.g., grain) removes another 4.3 Tg N/yr from the U.S.. Altogether, along with 5.4 Tg N/yr of atmospheric advection, these total Nr outputs out of the U.S. continental environment add up to about 14 Tg N/yr, leaving about 21 Tg N/yr unaccounted for. Of this amount, we estimate that 5 Tg N/yr are stored in soils, vegetation, and groundwater and, by difference, we estimate that about 16 Tg N/yr are denitrified to N_2. Denitrification, a process that microbially converts Nr to N_2 (as well as forming some N_2O) requires both a carbon source and anaerobic conditions, a situation that is found in wetlands, oxygen-depleted streams, rivers, and the hypolimnion of reservoirs (or their sediments), soils, and engineered denitrification systems. This process can be a major Nr sink in river basins. There are substantial uncertainties (+/- 50%) for estimated emission and deposition and terms that are arrived at by difference (e.g., atmospheric advection and denitrification) – especially those that involve NH_x. The Committee considered these uncertainties in developing the “Overarching Recommendations” of this report.

Current EPA Nr risk management and research programs

The parts of EPA most directly concerned with managing or conducting research on Nr are the Office of Air and Radiation, the Office of Water, and the Office of Research and Development (ORD). Over a dozen programs of EPA’s Office of Air and Radiation reduce risks from Nr. These programs and related activities include: National Ambient Air Quality Standards standard setting and implementation; emission standards for industrial stationary sources and area sources; the Acid Rain Program; the Clean Air Interstate Rule; and programs that focus on mobile source emissions. Programs designed to save energy, such as Energy Star, tend to reduce emissions of Nr as well. EPA’s Office of Water addresses Nr under both the Clean Water Act and the Safe Drinking Water Act through activities such as: criteria development and standard setting; total maximum daily load (TMDL) development; National Pollution Discharge Elimination System (NPDES) permits; infrastructure financing through the Drinking Water and Clean Water State Revolving Funds; watershed planning; wetlands preservation; and regulation of stormwater and runoff sources that include municipal separate storm sewer systems (MS4), and concentrated animal feeding operations (CAFOs). EPA’s Office of Research and Development aims to conduct leading-edge research and foster the sound use of science and technology in support of the Agency’s mission. The Office of Research and Development is well recognized for providing a scientific basis for the development of the National Ambient Air Quality Standards for NO_x and particulate matter (PM). The Office of Research and Development’s Ecosystem Services Research Program has been developed to identify and quantify the positive and negative impacts on ecosystem services resulting from changes in nitrogen loadings from major source categories. This research will support policy and management decisions in EPA’s Offices of Air and Radiation and Water.

EPA has brought a great variety of risk reduction tools to bear on Nr: conventional regulation and enforcement; cap and trade approaches; measurement, monitoring and place-based approaches; control technology development and verification; communication and education; intergovernmental and international cooperation; and voluntary approaches. The variety and breadth of EPA programs addressing Nr reflect the ubiquity of Nr in the environment, the historical single-medium regulatory approach, and the lack of a “silver bullet” for reducing risks from Nr.

Need for an integrated management strategy

The EPA programs discussed above (and the programs of EPA’s predecessor organizations) have been active in the management of Nr through efforts to: decrease or transform Nr in sewage; control NO_x to decrease photochemical smog and acid rain; control Nr inputs to coastal systems; control fine particulates in the atmosphere; and decrease Nr leaching and runoff from crop and animal production systems and developed lands. As beneficial as those efforts have been, they have focused on the specific problem without consideration of the interaction of a particular system with other systems downstream or downwind. Given the reality of the nitrogen cascade, this approach may result in short-term benefits for a particular system but may only temporarily delay larger-scale impacts on other systems. Thus there is a need to integrate N management programs, to ensure that efforts to lessen the problems caused by N in one area of the environment do not result in unintended problems in other areas.

Biofuels feedstock production provides a good example of the need for comprehensive and integrated assessment and management of Nr. Increasing corn production for ethanol has raised the prospect of increased Nr losses (i.e., transfer from fertilized land to water) and degraded water quality. The alternative of cellulosic based ethanol does not necessarily mitigate the potential for this negative externality. High yields of cellulosic materials also require N and the "marginal" land assumed for such production may be more susceptible to nutrient leakage. Another good example is provided in Chapter 4 of this report (Box 2). This example considers the water impacts of Nr in the Chesapeake Bay and shows that the total reduction of damage from excess Nr may rely nearly as much on stricter enforcement of the Clean Air Act as the Clean Water Act. This challenges the traditional approach to regulation, but it is a consequence of comprehensively examining Nr guided by the nitrogen cascade.

There can be many unintended consequences associated with a focus on managing one pollutant, even an integrated focus on various forms of N. For example, as further discussed in Chapter 4 and Appendix G of this report, numerous lakes, reservoirs, rivers, estuaries (e.g., the Gulf of Mexico), and fjords worldwide exhibit N and phosphorus (P) co-limitation, either simultaneously or in seasonally-shifting patterns. Therefore, strategies are needed to reduce both P and N inputs, and not all control practices will be effective for dual nutrient reduction. Synergistic effects on nutrient loss reductions can occur where combinations of control practices produce more or less than the sum of their individual reductions (U.S. EPA SAB, 2007). An integrated strategy should take this into consideration.

Objectives of the SAB Integrated Nitrogen Committee study

The EPA Science Advisory Board formed the Integrated Nitrogen Committee to assist EPA in its understanding and management of nitrogen-related air, land, and water pollution issues. In this report, the Committee has provided findings and recommendations addressing the following objectives. Assessment of the challenges and costs to EPA of implementing the recommendations is beyond the scope of the report.

1. Identify and analyze, from a scientific perspective, the problems Nr presents in the environment and the links among them.

To address this objective, the Committee used the nitrogen cascade framework to determine the major sources of newly created Nr in the U.S. (Figure ES-1). The flows of Nr within the food, fiber, feed and bioenergy production systems and developed lands in the U.S. were examined, paying special attention to the locations within each of these systems where Nr is lost to the environment. The same process was employed for fossil fuel energy production but, since all the Nr formed and released during energy production is lost to the environment, the Committee identified the important energy producing sectors that contribute to Nr emissions.

The Committee next examined the fate of the Nr lost to the environment, estimated the amount stored in different systems (e.g., forest soils) and tracked Nr as it is transferred from one environmental system (e.g., the atmosphere) to another (e.g., terrestrial and aquatic ecosystems).

Source and fate analyses set the stage for identifying the environmental and human health problems Nr presents, and the links among them. Using the nitrogen cascade, the Committee identified the impacts Nr has on people and ecosystem functions as it moves through each system. The Committee also addressed the alternative metrics that could be used, including the number of tons of specific forms of Nr, human health indicators and the economic damage cost, to assess incommensurable impacts due to environmental changes (e.g., acid deposition) vs. impacts due to losses of ecosystem services (e.g., loss of biodiversity), and trade-offs among Nr impacts.

2. Evaluate the contribution an integrated nitrogen management strategy[6] could make to environmental protection.

An integrated management strategy should take into account the contributions of all Nr sources, and all chemical species of Nr that adversely impact both human health and environmental systems. Further, an integrated strategy should ensure that solving one problem related to Nr does not exacerbate another problem or diminish ecosystem services that support societal demands. In short, the strategy should seek to achieve desirable benefits of Nr, while limiting adverse effects.

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[6] An integrated nitrogen management strategy takes a holistic approach for managing Nr. In the context of the nitrogen cascade, all Nr anthropogenic creation and destruction mechanisms and all Nr uses are recognized. The strategy should take account of synergies and trade-offs, to ensure that decreasing one problem related to nitrogen does not result in other unintended adverse environmental, economic and societal consequences. By identifying relative priorities, assessing cost effectiveness and risks, the strategy should seek to maximize the benefits of Nr, while limiting overall adverse effects.

To address this challenge, the Committee identified several actions that could be taken to better manage Nr in one environmental system and avoid unintended consequences in another. Examples of "integrative" management actions that could be taken are highlighted in that discussion.

3. Identify additional risk management options for EPA's consideration.

As further discussed below, the Committee has identified four major goals for management actions that collectively have the potential to decrease Nr losses to the environment by about 25 percent. Decreasing Nr emissions by these actions will result in further decreases in Nr-related impacts throughout the nitrogen cascade. The Committee has suggested several ways to attain these management goals including conservation measures, additional regulatory steps, voluntary actions, application of modern technologies, and end-of-pipe approaches. These are initial but significant actions; however, others should be taken once the recommended actions are completed and assessed, and further opportunities are explored in an adaptive management approach. Thus, the last sections of this report focus on a better understanding of Nr dynamics and impacts in the U.S. that could lead to more cost efficient management, balancing human and environmental needs.

4. Make recommendations to EPA concerning improvements in nitrogen research to support risk reduction.

In this report, the Committee has provided numerous recommendations for additional Nr research to support risk reduction activities. These research recommendations are discussed in various chapters of the report and are consolidated in the summary of findings and recommendations presented in Chapter 6.

Major Findings and Recommendations

Throughout the report there are boxes containing summary statements labeled "Findings." Attached to these findings are one or more specific "Recommendations" for actions that could be taken by EPA or other management authorities. In each case, the intent is to provide the scientific foundation regarding a specific Nr-relevant environmental issue and one or more recommendations by which EPA acting alone or in cooperation with other organizations could use currently available technology to decrease the amount of Nr lost to the U.S. environment. The findings and recommendations are consolidated in Chapter 6 of this report.

Overarching recommendations

Optimizing the benefits of Nr, and minimizing its impacts, will require an integrated nitrogen management strategy that involves action not only on the part of EPA, but also coordination with other federal agencies, the states, the private sector, universities, and the public, supported by a strong public outreach program. Therefore the Committee has also provided four overarching recommendations to assist EPA in its understanding and management of nitrogen-related air, land, and water pollution issues:

Overarching Recommendation 1: The Committee recommends an integrated approach to the management of Nr. This approach draws upon a combination of implementation mechanisms. Each mechanism must be appropriate to the nature of the problem at hand, be supported by critical research on decreasing the risks of excess Nr, and reflect an integrated policy that recognizes the complexities and tradeoffs associated with the nitrogen cascade. Management efforts at one point in the cascade may be more efficient and cost effective than control or intervention at another point. This is why understanding the nature and dynamics of the N cascade is critically important.

Overarching Recommendation 2: The framing of the reactive nitrogen cascade provides a means for tracking nitrogen as it changes form and passes through multiple ecosystems and media. This complexity requires the use of innovative management systems and regulatory structures to address the environmental and human health implications of the most damaging forms and quantities of Nr. It is difficult to create de novo fully effective regulations for such a complex system so we recommend utilizing adaptive management to continuously improve the effectiveness and lower the cost of implementation policies. This in turn will require a monitoring system that will provide feedback on the effectiveness of specific actions taken to lower fluxes and concentrations of Nr.

Overarching Recommendation 3: An intra-Agency Nr management task force within EPA is recommended to build on existing Nr research and management capabilities within the Agency. This task force should be aimed at increasing scientific understanding of: (1) Nr impacts on terrestrial and aquatic ecosystems, human health, and climate; (2) Nr-relevant monitoring requirements; and (3) the most efficient and cost-effective means by which to decrease various adverse impacts of Nr loads as they cascade through the environment.

Overarching Recommendation 4: Successful Nr management will require changes in the way EPA interacts with other agencies. Coordinated federal programs could better address Nr concerns and help ensure clear responsibilities for monitoring, modeling, researching and managing Nr in the environment. Thus, the Committee recommends that EPA convene an inter-agency Nr management task force. It is recommended that the members of this inter-agency task force include at least the following federal agencies: U.S. Department of Agriculture, U.S. Department of Energy, U.S. Department of Housing and Urban Development, U.S. Department of Transportation, National Oceanic and Atmospheric Administration, U.S. Geological Survey, U.S. Forest Service, and Federal Emergency Management Agency. The EPA Office of International and Tribal Affairs should work closely with the Department of State to ensure that EPA is aware of international efforts to control Nr and is developing national strategies that are compatible with international initiatives. Similar recommendations for coordination and joint action among and between agencies at both state and federal levels have been made in the National Research Council's recent reports on the Mississippi Basin (NRC, 2008b, 2009). These intra- and inter-agency Nr management task forces should take a systems approach to research, monitoring, and evaluation to inform public policy related to Nr management, and implement a systems approach to Nr management, as recommended by the Committee.

Summary of specific recommendations by study objective

The Committee's findings and recommendations corresponding to each of the four study objectives are summarized briefly below.

1. Identify and analyze, from a scientific perspective, the problems Nr presents in the environment and the links among them.

The Committee finds that uncertainty associated with rapid expansion of biofuels, losses of Nr from grasslands, forests, and urban areas, and the rate and extent of denitrification have created the need to measure, model, and report all forms of Nr consistently and accurately. Addressing this need will decrease uncertainty in the understanding of the fate of Nr that is introduced into the environment and lead to a better understanding of the impacts of excess Nr on the health of people and ecosystems. This should be accomplished through a coordinated effort among cognizant federal and state agencies, and universities.

In addition, the Committee recommends that EPA routinely and consistently account for the presence of Nr in the environment in forms appropriate to the medium in which they occur (air, land, and water) and that accounting documents be produced and published periodically (for example, in a fashion similar to National Atmospheric Deposition Program summary reports). The Committee understands that such an undertaking will require substantial resources, and encourages the Agency to develop and strengthen partnerships with appropriate federal and state agencies, and private sector organizations, with parallel interests in advancing the necessary underlying science of Nr creation, transport and transformation, impacts, and management.

2. Evaluate the contribution an integrated nitrogen management strategy could make to environmental protection.

The Committee finds that effective management of Nr in the environment must recognize the existence of tradeoffs across a number of impact categories involving the cycling of nitrogen and other elements. In addition, an integrated multi-media approach to monitoring Nr is needed.

In that regard, the Committee recommends that:

1. EPA should develop a uniform assessment and management framework that considers the effects of Nr loading over a range of scales reflecting ecosystem, watershed, and regional levels. The framework should include all inputs related to atmospheric and riverine delivery of Nr to estuaries, their comprehensive effects on marine eutrophication dynamics, and their potential for management.
2. EPA should examine the full range of traditional and ecosystem response categories, including economic and ecosystem services, as a basis for expressing Nr impacts in the environment, and for building better understanding and support for integrated management efforts.

3. Identify additional risk management options for EPA's consideration.

The Committee finds that a number of risk management actions should be considered to reduce Nr loading and transfer to the environment. These include farm-level improvements in manure management, actions to reduce atmospheric emissions of Nr, and interventions to control Nr in water management programs. As an example, the Committee recommends that EPA should reexamine the criteria pollutant "oxides of nitrogen" and the indicator species NO_2 and consider supplementing this with NH_x and NO_y as indicators of chemically reactive nitrogen (Nr without N_2O).

4. Make recommendations to EPA concerning improvements in nitrogen research to support risk reduction.

The Committee finds that research is needed in a number of areas to support Nr risk reduction activities. These areas include research to advance the understanding of: the quantity and fate of Nr applied to major crops; how to accelerate crop yields while increasing N fertilizer uptake efficiency; agricultural emissions of forms of Nr; atmospheric deposition of Nr; and the potential for amplification of Nr-related climate impacts.

Four recommended management actions

Consistent with the overarching and specific recommendations noted above, the Committee identified four management actions that could be undertaken in the near term by applying existing proven science and technology and determined how those actions could contribute to the reduction of excess Nr in the environment.

1. The Committee estimates that if EPA were to expand its NO_x control efforts for emissions of mobile sources and power plants, a **2.0 Tg N/yr** decrease in the generation of reactive nitrogen could be achieved. Such changes can be effected by applying existing, proven technology. Emissions from many point sources are controlled with low-NO_x burners or NO_x reduction. Such equipment should also be installed on industrial boilers and the remaining, uncontrolled power plants. NO_x controls for modern on-road vehicles are effective and these technologies should be applied to off-road vehicles, locomotives, ships and other devices with internal combustion engines.
2. The Committee estimates that excess flows of Nr into streams, rivers, and coastal systems can be decreased by approximately 20% (approximately **1 Tg N/yr**) through improved landscape management and without undue disruption to agricultural production. This would include activities such as using large-scale wetland creation and restoration to provide needed ecosystem services of Nr retention and conversion as well as matching cropping systems and intensity of Nr use to land characteristics. Improved tile-drainage systems and riparian buffers on cropland, and implementing stormwater and non-point source management practices (e.g., EPA permitting and funding programs) are important components. In addition, the Committee estimates that crop N-uptake efficiencies can be increased by up to 25% over current practices through a combination of knowledge-based practices and advances in fertilizer technology (such as controlled release and inhibition of nitrification). Crop output can be increased while decreasing total Nr by up to 20% of applied artificial Nr, amounting to ~**2.4 Tg N/yr** below current amounts of Nr additions to the environment. These are appropriate actions that could be taken with today's available technologies and further progress is possible.
3. The Committee estimates that livestock-derived NH_3 emissions can be decreased by 30% (a decrease of **0.5 Tg N/yr**) by a combination of BMPs and engineered solutions. This is expected to decrease $PM_{2.5}$ by approximately 0.3 micrograms per cubic meter (2.5%), and improve health of ecosystems by achieving progress towards critical load recommendations. Additionally we estimate that NH_3 emissions derived from fertilizer applications can be decreased by 20% (decrease by approximately **0.2 Tg N/yr**), through BMPs that focus on improvements related to application rate, timing, and placement.
4. The Committee recommends that a high priority be assigned to increasing funding for nutrient management. We estimate that adequate financial support for sewage treatment infrastructure upgrades to remove nutrients could decrease Nr emissions by between **0.5 and 0.8 Tg N/yr**. Additional Nr management from eligible stormwater and nonpoint sources could be accomplished through increased support.

Implementing these suggestions will decrease the amount of Nr introduced into the United States by about 25%, which will similarly decrease the amount of Nr lost to the atmosphere, soils and waters. The Committee believes that these represent realistic and attainable near-term outcomes, however further reductions are undoubtedly needed for many N-sensitive ecosystems and to ensure that health-related standards are maintained.

Reactive Nitrogen in the United States

AN ANALYSIS OF INPUTS, FLOWS, CONSEQUENCES, AND MANAGEMENT OPTIONS

1

Introduction

1.1. Overview of the Problem – Impacts of Excess Nr on Human Health and the Environment

Nitrogen is an essential nutrient that governs the growth and reproduction of living organisms. Reactive nitrogen (Nr), in contrast to non-reactive gaseous N_2, includes all biologically active, chemically reactive, and radiatively active nitrogen (N) compounds in the atmosphere and biosphere of the Earth. Anthropogenic creation of Nr provides essential benefits for humans – first and foremost in meeting human dietary needs. In fact, a large proportion of the human population of the earth could not be sustained if synthetic nitrogen fertilizers did not augment food production significantly all over the world. However, excess releases of Nr to the environment from human activities such as fossil fuel combustion and agriculture are a major cause of air and water quality degradation that has been linked to significant impacts on human and ecosystem health.

Reactive nitrogen (Nr) includes inorganic chemically reduced forms of N (NH_x) [e.g., ammonia (NH_3) and ammonium ion (NH_4+)], inorganic chemically oxidized forms of N [e.g., nitrogen oxides (NO_x), nitric acid (HNO_3), nitrous oxide (N_2O), N_2O_5, HONO, peroxy acetyl compounds such as peroxyacytyl nitrate (PAN), and nitrate ion (NO_3-)], as well as organic compounds (e.g., urea, amines, amino acids, and proteins).

The negative consequences of Nr flux in the U.S. environment include increases in photochemical smog and atmospheric particulate matter ($PM_{2.5}$), decreases in atmospheric visibility, both increases and decrease in productivity of grasslands and forests, acidification of soils and freshwaters, accelerating estuarine and coastal eutrophication, increases in the emission of greenhouse gases (GHG) to the atmosphere, and decreases in stratospheric ozone concentrations. Most of these changes in environmental conditions lead to a variety of negative impacts on both ecosystem and human health (Johnson and Siccama, 1983; Heck et al., 1984; Paerl, 1988; MacKenzie and El-Ashry, 1990; ECOHAB, 1995; Bricker et al., 1999; Rabalais et al., 1999; NRC, 2000; Mitsch et al., 2001; Fenn et al., 2003; Ezzati et al., 2004; Mokad et al., 2004; Verity et al., 2006; U.S. EPA Clean Air Scientific Advisory Committee, 2008; U.S. EPA SAB, 2008; Bobbink et al., 2010). In light of the magnitude of the human alteration of the nitrogen cycle, and the resulting negative consequences on humans and ecosystems, the National Academy of Engineering has identified management of the nitrogen as one of the "grand challenges" facing this country (National Academy of Engineering, 2008).

1.2. The Nitrogen Cascade – Nr Loading, Cycling, and Exposure

Approximately 78% of the atmosphere is diatomic nitrogen (N_2), which is unavailable to most organisms because of the strength of the triple bond that holds the two N atoms together. Over evolutionary history, only a limited number of species of bacteria and archaea have evolved the ability to convert N_2 to Nr via biological N fixation. Thus, even with adaptations to use N efficiently, many ecosystems of the world are limited by N.

Anthropogenic creation of Nr

Nitrogen limitation of ecosystem production has driven humans to use increasingly sophisticated and energy-intensive measures to obtain Nr to sustain food production and to produce other commodities (e.g., nylon, explosives). In pre-history, hunters and gatherers harvested food from natural stocks. With the advent of agriculture, local sources of Nr were used (soil stocks, crop residue, and manures) to increase productivity of landscapes. In the nineteenth century, long-range transport of Nr to sustain food production increased with the shipment of bird guano from the Pacific Islands and nitrates from South America to Europe and other locations. By the beginning of the twentieth century, these sources were not sufficient to sustain the growing global population requirements for food.

This deficiency led to what has been called one of the world's most important discoveries – how to extract N_2 from the atmosphere and convert it to ammonia (NH_3) – called the Haber-Bosch process (Smil, 2001; Erisman et al., 2008). Today, this process and cultivation-induced biological N fixation (C-BNF) introduce over 140 teragrams (Tg) of N per year (hereafter expressed as Tg N/yr) into the global environment to increase food production (Galloway et al., 2008). Another 23 Tg N/yr are introduced by the Haber-Bosch process for the chemical industry, and 25 Tg N/yr are introduced via the combustion of fossil fuels (Galloway et al., 2008).

The total global anthropogenic Nr creation rate is ~190 Tg N/yr (2005), substantially larger than the median of estimates for Nr creation by natural terrestrial processes (~100 Tg N/yr) (Galloway et al., 2008). The fact that humans are more effective than nature in Nr creation means that on average, humans are less reliant on natural sources of Nr. However, with global commodity stocks running at a 58-day supply and food prices increasing dramatically, the challenge is to increase the nutrient use efficiency of Nr in agricultural systems while maintaining or increasing yields (USDA, ERS/World Agricultural Outlook Board, July 11, 2008. World Agricultural Supply and Demand Estimates).

There are large regional disparities in Nr creation rates on both absolute and per capita bases. Total Nr creation is larger in Asia than in any other region. Per capita Nr creation is largest in North America and Europe. Humans also redistribute large amounts of Nr among countries or regions of the world through exports of fertilizers, feed grains, and fossil fuels. Nevertheless, there are large regions of the world with populations approaching one billion, where there is malnutrition in part due to a lack of available Nr to sustain crop production.

The introduction of Nr into most regions of the United States by humans has greatly increased food availability. However, since essentially all the Nr created for food production and by fossil fuel combustion is lost to the environment, it has also greatly increased the contribution of Nr to a wide variety of environmental problems. Most plants, animals, and microorganisms are adapted to efficiently use and retain Nr. Addition of Nr to most ecosystems may first lead to increased uptake, growth, and storage – and hence to increased biomass, including food or fiber production. However, further addition of Nr in excessive amounts often leads to imbalances in the movement of Nr among reservoirs and potential losses[7] to the environment in the form of air emission or water discharges into other ecosystems where Nr may disrupt ecosystem functions and have a negative impact on resources. In essence, the assimilative capacity of the ecosystem may be insufficient to benefit from increases in Nr without disruptive changes.

These changes, which impact air, land, water and the balance of life in an interrelated fashion, are often referred to as a cascade of effects from excess Nr[8] or the "nitrogen cascade" (Figure 1). Unlike other element-based pollution problems, the N cascade links the negative impacts, where one N-containing molecule can in sequence contribute to all the environmental issues mentioned above.

The nitrogen cascade has three dimensions: biogeochemical, alterations in the environment, and human and ecosystem consequences.

The "biogeochemical" dimension of the nitrogen cascade involves: Nr creation from N_2 as a consequence of chemical, food and energy production; Nr use in food and chemical production; Nr losses to the environment; changes in Nr species residence times in environmental reservoirs; Nr transfers among reservoirs; and Nr conversion back to N_2. Alterations to the environment then result from increased Nr levels in the environment. These alterations have negative consequences for ecosystem and human health at local, regional, national and global scales. Because nitrogen is a critical resource and also a contributor to many of the environmental concerns facing the U.S. today, it is imperative to understand how human action has altered N cycling in the U.S., and the consequences of those alterations on people and ecosystems. The overarching question is, how do we protect and sustain ecosystems that provide multiple benefits to society while also providing the interconnected material, food and energy required by society?

Nr inputs to the nation and the world have been increasing, largely due to human activities associated with food production and fossil fuel combustion. Despite the obvious benefits of a plentiful supply of food and energy, the adverse consequences associated with the accumulation of Nr in the environment are large, with implications for human health and the environment.

The greater the inputs of Nr to the landscape, the greater the potential for negative effects, caused by greenhouse gas (GHG) production, ground level ozone, acid deposition, and Nr overload that can contribute to climate change, degradation of soils and vegetation, acidification of streams, lakes and rivers, estuarine and coastal eutrophication, hypoxia and habitat loss.

The growing nature of the Nr problem, and the adverse and intertwined consequences associated with Nr inputs to air, land, and water as exhibited in the N cascade underscore the need for researchers and managers to explore integrated strategies that minimize N inputs, maximize its use efficiency, promote Nr removal processes and protect humans and natural resources.

The concept of the nitrogen cascade highlights that once a new Nr molecule is created, it can, in sequence, travel throughout the environment contributing to major environmental problems (Galloway et al., 2003). The adaptation of the cascade in Figure 1 was developed by the SAB Integrated Nitrogen Committee (INC) to provide a context for considering nitrogen-related issues and ecosystem effects in the U.S. To consider the cascading effects of Nr in the U.S., we examined the various atmospheric, terrestrial, and aquatic environmental

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[7] In the context of this report, "losses" refers to transfers among systems and not the conversion of Nr to N_2. Whenever N_2 formation is discussed, it is explicitly stated.

[8] Excess reactive nitrogen (Nr) is defined as the amount of Nr that is present in, or introduced into, an environmental system (e.g., Nr inputs to the atmosphere, Nr inputs to grasslands and forests, Nr inputs to estuaries) from anthropogenic sources that is not incorporated into agricultural and other biological products (e.g., food, feed, fuel, and fiber), or stored in long-term storage pools (e.g., cropland soils).Thresholds are used to determine the amount of excess Nr that causes negative effects on ecosystem services and functions, and human health. Thresholds vary by metric (e.g., concentration, loading, etc) and depend on the environmental system (e.g., atmosphere, forest). Examples for specific thresholds are given later in the report in relevant sections.

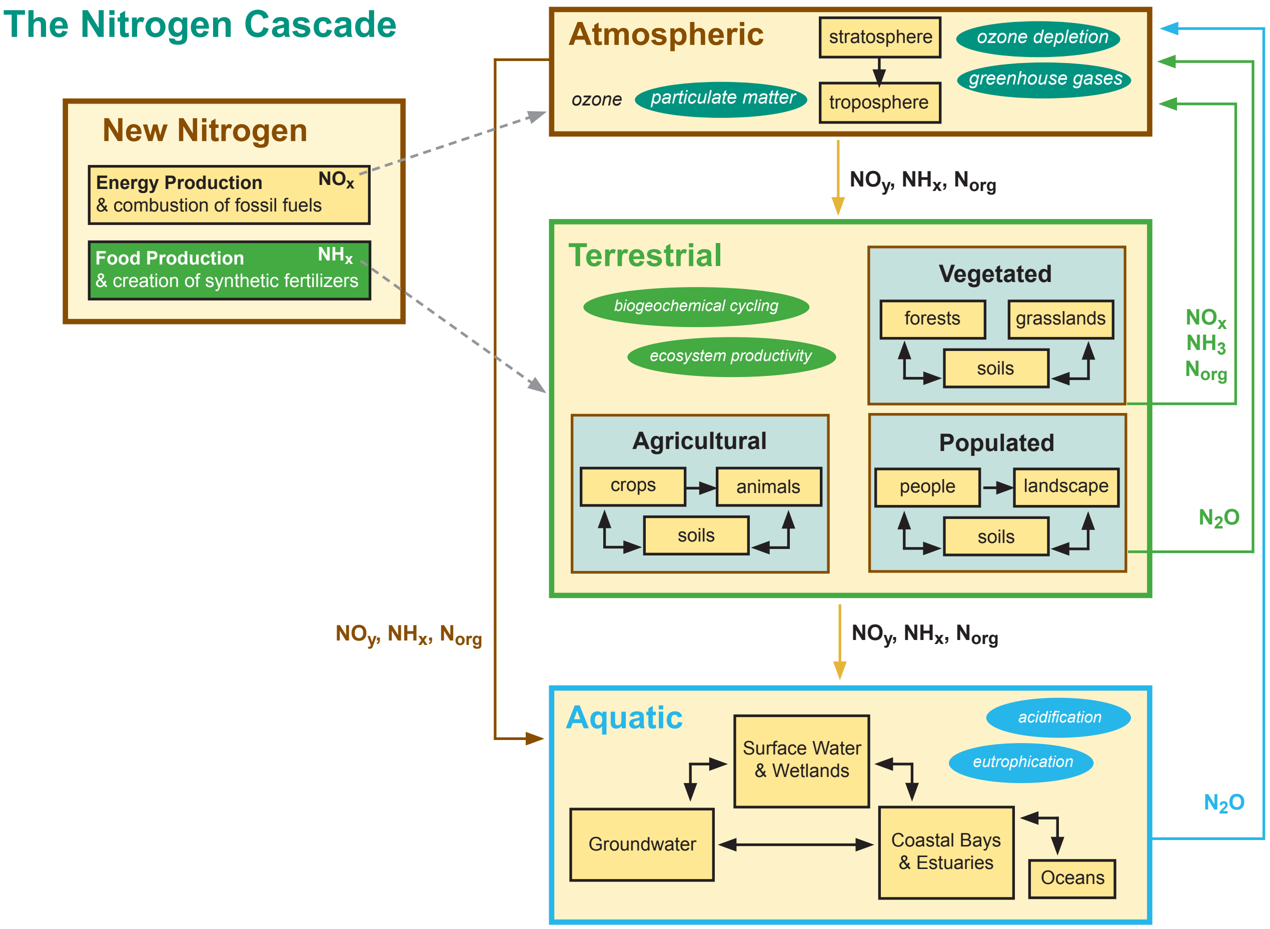

Figure 1: The nitrogen cascade

systems where Nr is stored, and the magnitudes of the various flows of N to, from, and within them. The nitrogen cascade concept implies the cycling of Nr among these systems. The process of denitrification is the only mechanism by which Nr is converted to chemically inert N_2, "closing" the continuous cycle (Figure 1 shows only flows of reactive nitrogen, not N_2). Denitrification can occur in any of the indicated reservoirs except the atmosphere.

The "new" N box in Figure 1 depicts the two primary anthropogenic sources by which Nr originates – energy production and food production – and where Nr from these sources enter ecosystems. Energy production includes both fossil fuel and biofuel combustion. Food production includes N fertilizer produced in the U.S., cultivation-induced biological N (C-BNF) in the U.S., production of animals and crops in the U.S. for human consumption, and imports of N-containing fertilizer, grain and meat to the U.S.

The atmospheric system box in Figure 1 indicates that tropospheric concentrations of both ozone and particulate matter are increased due to emissions of nitrogen oxides[9] (NO_x) to the atmosphere. The ovals illustrate that the increase in N_2O concentrations, in turn, contribute to the greenhouse effect in the troposphere and to ozone depletion in the stratosphere. Except for N_2O, there is limited Nr storage in the atmosphere. Losses of Nr from the atmospheric system include total oxidized nitrogen[10] (NO_y), reduced nitrogen[11] (NH_x), and organic nitrogen (N_{org}) deposition to terrestrial and aquatic ecosystems of the earth's surface. There is little potential for conversion of Nr to N_2 via denitrification in air. However, once airborne deposition of Nr occurs it will be subject to denitrification pathways via soil and water.

The terrestrial system box in Figure 1 depicts that Nr enters agricultural lands via food production and is introduced to the entire terrestrial landscape via atmospheric deposition. Within agricultural regions there is cycling among soils, crops and animals, and then a transfer of Nr as food to populated regions, from which there are Nr losses to the environment (e.g., sewage,

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[9] NO_x (oxides of nitrogen) includes NO + NO_2

[10] NO_y (total oxidized nitrogen) includes NO, NO_2, NO_3, N_2O_5, HONO, HNO_3, NO_3^-, PAN and other organo-nitrates, $RONO_2$

[11] NH_x (reduced nitrogen) includes NH_3 + NH_4

landfills). The ovals showing ecosystem productivity and biogeochemical cycling reflect that Nr is actively transported and transformed within the terrestrial system, and that as a consequence there are significant impacts on ecosystem productivity due to fertilization and acidification, often with resulting losses of biodiversity. There is ample opportunity for Nr storage in both biomass and soils. Losses of Nr from this system occur by leaching and runoff of NO_y, NH_x and N_{org} to aquatic ecosystems and by emissions to the atmospheric system as NO_x, NH_3, N_{org}, and N_2O. There is potential for conversion of Nr to N_2 via denitrification in the terrestrial system.

The aquatic system box in Figure 1 shows that Nr is introduced via leaching and runoff from terrestrial ecosystems and via deposition from atmospheric ecosystems. Connected with the hydrological cycle, there are Nr fluxes downstream with ultimate transport to coastal systems. Within the aquatic system, the ovals highlight two significant impacts of waterborne Nr acidification of freshwaters and eutrophication of fresh and coastal waters. Except for Nr accumulation in groundwater reservoirs, there is limited Nr storage within the hydrosphere. Losses of Nr from the aquatic system are primarily via N_2O emissions to the atmospheric system. There is a very large potential for conversion of Nr to N_2 via denitrification in water and wetlands.

NO_y, NH_x and N_2O are all components of Nr, but a fundamental difference is that the NO_y and NH_x are rapidly transferred from the atmosphere to receiving ecosystems due to a short atmospheric residence time ($\leq$ 10 days) where they continue to contribute to the N cascade (Galloway et al., 2004). Because of its longer residence time (~100 years) however, N_2O remains in the troposphere where it contributes to climate change, until it is transferred to the stratosphere, where it contributes to ozone depletion (Galloway et al., 2004).

1.3. EPA Activities to Manage Risks Posed by Nr

EPA activities to manage the risks posed by reactive nitrogen can be linked to the Agency's broad strategic goals. EPA's mission is to protect human health and the environment. In achieving this mission, EPA is accountable for addressing five goals given in the *2006 – 2011 EPA Strategic Plan* (U.S. EPA, 2006d):

1. Clean air and global climate change
2. Clean and safe water
3. Land preservation and restoration
4. Healthy communities and ecosystems
5. Compliance and environmental stewardship

The *Strategic Plan* includes targets for reducing risk from N. EPA's *Report on the Environment* (U.S. EPA, 2008c), provides "data on environmental trends," to determine whether or not EPA is on track to meet its targets and goals. EPA is responsible and accountable for reducing at least some risks from Nr.

As previously discussed, the principal mechanisms for Nr removal from circulation in the environment are complete denitrification (re-conversion of Nr back to non-reactive gaseous N_2), and storage in long-term reservoirs (e.g., soils, sediments, and woody biomass). In some cases, it may be possible to capture Nr emissions or discharges and deliver them to food or fiber production areas where there are nitrogen deficiencies. However, as previously noted, major challenges in the management of the N cycle are how to decrease creation of Nr while still meeting societal needs, promote denitrification of excess Nr (without producing N_2O), and improve the efficiency of use and reuse of excess Nr in a cost-effective manner. Solving these challenges will result in less Nr accumulation.

The parts of EPA most directly concerned with managing or conducting research on Nr are the Office of Air and Radiation, the Office of Water, and the Office of Research and Development. Programs designed to save energy, such as Energy Star, tend to reduce emissions of Nr as well. In over a dozen programs, EPA's Office of Air and Radiation reduces risks from Nr. These programs and related activities include:

- National Ambient Air Quality Standards (NAAQS) standard setting and implementation;
- Emission standards for industrial stationary sources and area sources
- Acid Rain Program
- Clean Air Interstate Rule
- Programs that focus on mobile source emissions

EPA's Office of Water addresses Nr under both the Clean Water Act and the Safe Drinking Water Act through activities such as:

- Criteria development and standard setting
- Total maximum daily load (TMDL) development
- National Pollution Discharge Elimination System (NPDES) permits
- Watershed planning
- Wetlands preservation
- Regulation of concentrated animal feeding operations (CAFOs).

EPA's Office of Research and Development aims to conduct leading-edge research and foster the sound use of science and technology in support of EPA's mission. The Office of Research and Development is well recognized for providing a scientific basis for the development of the National Ambient Air Quality Standards for NO_x and particulate matter (PM). The Office of Research and Development's Ecosystem Services Research Program has been developed to identify and quantify the positive and negative impacts on ecosystem services resulting from changes in nitrogen loadings from major source categories to support policy and management decisions in EPA's Offices of Air and Radiation and Water.

EPA has brought a great variety of risk reduction tools to bear on reactive N:

- Conventional regulation and enforcement
- Cap and trade approaches
- Measurement, monitoring and place-based approaches
- Control technology development and verification
- Communication and education
- Intergovernmental and international cooperation
- Voluntary approaches

The variety and breadth of EPA programs addressing Nr reflect the ubiquity of Nr in the environment, the historical single medium regulatory approach, and the lack of a silver bullet for reducing risks from Nr.

Need for an Integrated Nitrogen Management Strategy

The EPA programs discussed above (and the programs of EPA's predecessor organizations) have been active in the management of Nr through efforts to: decrease the Nr amount in sewage, control NO_x to decrease photochemical smog and acid rain, control Nr inputs to coastal systems, control fine particulates in the atmosphere, and decrease Nr leaching and runoff from crop and animal production systems. As beneficial as those efforts have been, they have focused on the specific problem without consideration of the interaction of their particular system with other systems downstream or downwind. Given the reality of the nitrogen cascade, this approach may result in short-term benefits for a particular system but will also likely only temporarily delay larger-scale impacts on other systems. Thus, there is a need to integrate N management programs, to ensure that efforts to lessen the problems caused by N in one area of the environment do not result in unintended problems in other areas.

Biofuels feedstock production is a good example of this. Increasing corn production for ethanol raised the prospect of increased Nr losses and degraded water quality. The alternative of cellulosic based ethanol does not necessarily mitigate the potential for this negative externality. High yields of cellulosic materials also require N and the "marginal" land assumed for such production may be more susceptible to nutrient leakage (NRC, 2008a).

In addition, there can be unintended consequences associated with a focus on one pollutant, even an integrated focus on various forms of nitrogen. For example, as further discussed in Chapter 4 and Appendix G of this report, numerous lakes, reservoirs, rivers, estuaries (e.g., the Gulf of Mexico), and fjords worldwide exhibit N and P co-limitation, either simultaneously or in seasonally-shifting patterns. Therefore, strategies are needed to reduce both P and N inputs, and not all control practices will be effective for dual nutrient reduction. There can be synergistic effects on nutrient loss reductions where combinations of control practices can produce more or less than the sum of their individual reductions (U.S. EPA SAB, 2007) and an integrated strategy should take this into consideration.

1.4. SAB Integrated Nitrogen Committee Study Objectives

The EPA Science Advisory Board has previously provided advice concerning management of nitrogenous compounds as well as integrated environmental decision making. In 1973, the Science Advisory Board issued a report *Nitrogenous Compounds in the Environment* (U.S. EPA SAB, 1973). The report addressed sources and effects of nitrogenous compounds, including those from air emissions, animal wastes, crop agriculture, industrial processes, and solid wastes. The SAB concluded that, "At present, all known trends appear to be ones that can be managed and kept within control, if appropriate steps are taken now," and provided recommendations relating to Nr research and control. In its 2000 report, *Toward Integrated Environmental Decision-Making* (U.S. EPA SAB, 2000) the SAB articulated a framework for integrated environmental decision-making. In that report, the SAB noted that the three-phased structure of the framework (problem formulation, analysis and decision-making, followed by implementation and evaluation) "belies the complexities involved in putting the concept of integrated decision-making into practice." The SAB's interests in N science and integrated environmental protection converged in 2007, when the SAB identified integrated N research and control strategies as an important issue facing the Agency and formed the Integrated Nitrogen Committee (the Committee) to conduct this study.

The Committee was charged by the Science Advisory Board to address the following four objectives:

1. Identify and analyze, from a scientific perspective, the problems reactive nitrogen presents in the environment and the links among them;
2. Evaluate the contribution an integrated nitrogen management strategy[12] could make to environmental protection;

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[12] An integrated nitrogen management strategy takes a holistic approach for managing Nr. In the context of the nitrogen cascade, all Nr anthropogenic creation and destruction mechanisms and all Nr uses are recognized. The strategy should take account of synergies and trade-offs, to ensure that decreasing one problem related to nitrogen does not result in other unintended adverse environmental, economic and societal consequences. By identifying relative priorities, assessing cost-effectiveness and risks, the strategy should seek to maximize the benefits of Nr, while limiting overall adverse effects.

3. Identify additional risk management options for EPA's consideration; and
4. Make recommendations to EPA concerning improvements in nitrogen research to support risk reduction.

In this report the Committee has provided findings and recommendations addressing the study objectives. We recognize that there will be challenges and costs associated with the pursuit of the recommended management strategies, and that EPA requires statutory authority to take regulatory action. However, assessment of the challenges and costs to the Agency of implementing the recommendations is beyond the scope of this report.

1.5. Study Approach and Structure of the Report

To address the four objectives of this study, the Committee completed the following activities:

1. The Committee used the nitrogen cascade framework to determine the major sources of newly created Nr in the U.S. The flows of Nr within the food, fiber, feed, and bioenergy production systems of the U.S. were examined, paying special attention to the locations in each of these systems where Nr is lost to the environment. The same process was employed for energy production but, since all of the Nr formed during energy production is lost to the environment, the Committee identified the important energy producing sectors that contribute to Nr formation.
2. The Committee examined the fate of Nr lost to the environment, estimated the amount stored in different systems (e.g., forest soils), and tracked Nr as it is transferred from one environmental system (e.g., the atmosphere) to another (e.g., terrestrial and aquatic ecosystems).
3. Using the nitrogen cascade, the Committee identified the impacts Nr has on people and ecosystem functions as it moves through different systems.
4. The Committee identified actions that could be taken based on available science and management practices to improve the integrative management of N. The Committee suggested ways in which each of these actions could be accomplished and estimated that together they could decrease Nr losses to the environment by about 25%.
5. The Committee identified research needed to improve the scientific foundation to support specific Nr risk reduction activities.

Four public meetings were held during the course of the study and briefings were presented to the Committee by: EPA's Office of Air and Radiation, Office of International Affairs, and Office of Water; the U.S. Department of Agriculture's Agricultural Research Service, Cooperative State Research, Extension and Education Service, and the Economic Research Service; and external organizations such as the Energy Research Centre of the Netherlands, Environmental Defense Fund, International Plant Nutrition Institute, Iowa State University, LiveFuels, and the Soil and Water Conservation Society.

Additionally, the Committee invited scientists and managers from EPA, other federal agencies, states and localities, academia, non-governmental organizations, and the private sector to participate in an October 20-22, 2008 workshop and meeting on Nitrogen Risk Management Integration. The purpose of the workshop was to receive public input on several subjects: the Committee's preliminary assessment of Nr problems, consequences, and remedies, with emphasis on risk reduction; the Committee's quantitative estimates of attainable Nr reductions; and mechanisms whereby the Nr strategy might be enacted. The Committee took this public input into consideration as it developed this report.

Structure of the report

This report contains six chapters. The report was developed for a multifaceted audience of scientists and policy makers and therefore the level of detail varies in different sections of the document. The introductory chapter provides an overview of problems caused by excess reactive nitrogen and describes the study objectives and approach. Chapters 2-6 discuss how the Committee has addressed the four study objectives and present specific findings and recommendations. The findings and recommendations corresponding to each of the study objectives are consolidated in Chapter 6.

- Study objective 1 (identification and analysis of the problems nitrogen presents in the environment and linkages among these problems) is addressed in Chapters 2 and 3. Chapter 2 focuses on the sources, transfer, and transformation of reactive nitrogen in environmental systems. Chapter 3 describes the impacts of reactive nitrogen on aquatic, atmospheric, and terrestrial ecosystems.
- Study objective 2 (evaluation of the contribution an integrated nitrogen management strategy could make to environmental protection) is addressed in Chapters 4 and 5. Chapter 4 reviews the implications for risk reduction strategies for reactive nitrogen. Chapter 5 discusses integrated risk reduction strategies.
- Study objective 3 (identification of additional risk management options for EPA's consideration) is addressed in Chapters 5 and 6. In Chapter 6, the Committee identifies specific management goals for reducing the loss of reactive nitrogen to the environment. The Committee believes that these represent realistic near-term management goals that can be attained using current technology. However, the Committee emphasizes that further reduction beyond

these goals will be needed to protect many N-sensitive ecosystems and to ensure that health-related standards are maintained.

The Committee finds that the management goals could be attained by conservation measures, additional regulation, and application of modern technologies.

- Study objective 4 (recommendation of improvements in reactive nitrogen research to support risk reduction) is addressed in all of the report chapters and Chapter 6 contains a section describing the need for a comprehensive program to monitor Nr in the environment.

Throughout this report there are boxes containing summary statements labeled “Findings.” Attached to these findings are one or more specific “Recommendations” for actions that could be taken by EPA or other management authorities.

2

Sources, Transfer, and Transformation of Nr in Environmental Systems

The Committee was charged with identifying and analyzing, from a scientific perspective, the problems Nr presents in the environment and the links among them. This chapter addresses two aspects of the Committee's work. The first aspect is the introduction of Nr into U.S. environmental systems from fossil fuel combustion and from food production, and the second aspect is the fate of Nr after it is emitted to the atmosphere by fossil fuel combustion or lost to the air, water and soils from agricultural production systems. The Nr budgets and calculations for the U.S. exclude Alaska and Hawaii because data were not available for these areas. Most of the Nr introduced into the environment comes from the Haber-Boesch process. Haber-Boesch contributes three to four times the Nr introduced from fossil fuel combustion and most of the Haber-Boesch introduction of Nr comes through agriculture.

2.1. Nr Flux in the Environment

As stated previously, although N is a major required nutrient that governs growth and reproduction of living organisms, Nr losses to the environment from human sources have a profound effect on air, water and soil quality. Human consumption of energy to sustain economic development results in emissions of NO_x to the atmosphere via fossil fuel combustion. Consumption of food to meet nutritional requirements of a growing population results in agricultural emissions of NH_3, urban and industrial emissions of NO_x and N_2O, and losses of NO_3^- and other N compounds to water bodies due to leaching and runoff. Once released into the atmosphere by either human or natural processes, these Nr compounds undergo transformation through atmospheric reactions (e.g., gas-to-particle conversion), transport associated with wind, and finally wet and dry deposition. Reactive nitrogen lost from agricultural and peopled systems can enter groundwater, streams, lakes, estuaries, and coastal waters where the Nr can also undergo transformation mediated by a wide range of biotic and abiotic processes. The introduction of Nr into agroecosystems provides much of the world's food. The losses of Nr to the environment throughout the food production process and during fossil fuel combustion contribute to many of the major environmental problems of today. The impacts of Nr on humans and ecosystems are discussed in the Executive Summary and Chapter 3 of this report.

Some key issues concerning management of Nr in the U.S. environment are the introduction of new Nr by imports, fertilizer production, cultivation-induced biological nitrogen fixation (C-BNF), and fossil fuel combustion. Other important issues are the distribution of Nr within agricultural systems and populated systems and redistribution of Nr through losses from those systems to the environment (Figure 1). National-level values for Nr fluxes are displayed in Table 1. Fluxes that represent the introduction of new Nr into the U.S. are marked with an asterisk and illustrated in Figure 1. In specific sections of this report these values have been used to more clearly determine the flux and fate of Nr in the U.S. Figure 2 Illustrates sources of Nr introduced into the United States.

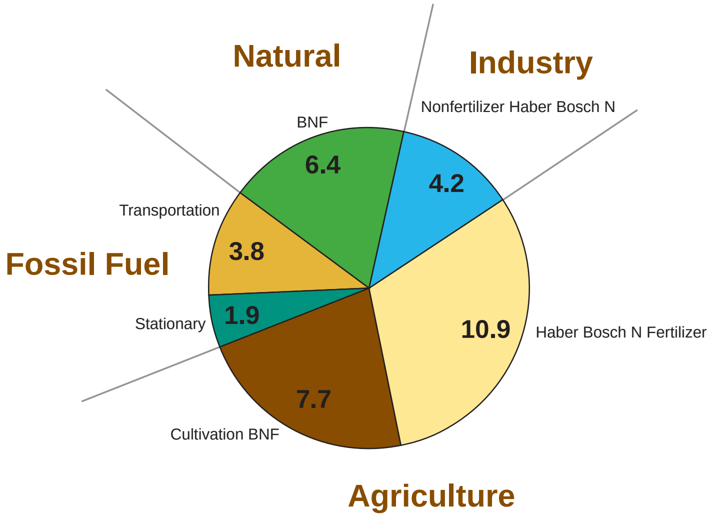

Figure 2: Sources of reactive nitrogen (Nr) introduced into the United States in 2002 (Tg N/yr).

Table 1: Nr fluxes for the United States, Tg N in 2002.[a]

Nr inputs to the *Atmospheric* environmental system	Tg N/yr*	%
N_2O-N emissions[1]	0.8	8
Agriculture - livestock (manure) N_2O-N	0.03	
Agriculture – soil management N_2O-N	0.5	
Agriculture - field burning agricultural residues	0.001	
Fossil fuel combustion - transportation*	0.1	
Miscellaneous	0.1	
NH_x-N emissions[2]	3.1	31
Agriculture: livestock NH_3-N	1.6	
Agriculture: fertilizer NH_3-N	0.9	
Agriculture: other NH_3-N	0.1	
Fossil fuel combustion – transportation *	0.2	
Fossil fuel combustion - utility & industry *	0.03	
Other combustion	0.2	
Miscellaneous	0.1	
NO_x-N emissions[2]	6.2	61
Biogenic from soils	0.3	
Fossil fuel combustion – transportation *	3.5	
Fossil fuel combustion - utility & industry *	1.9	
Other combustion	0.4	
Miscellaneous	0.2	
Total *Atmospheric* inputs	**10.0**	**100**
Nr inputs to the *Terrestrial* environmental system		
Atmospheric N deposition[b]	6.9	16
Organic N[3]	2.1	
Inorganic NO_y-N[4]	2.7	
Inorganic-NH_x-N[4]	2.1	
N fixation in cultivated croplands * [5]	7.7	18
Soybeans*	3.3	
Alfalfa*	2.1	
Other leguminous hay *	1.8	
Pasture*	0.5	
Dry beans, peas, lentils *	0.1	
N fixation in non-cultivated vegetation * [6]	6.4	15
N import in commodities *[7]	0.2	0.3
Synthetic N *[8]	15.1	35
Fertilizer use on farms & non-farms	10.9	
Non-fertilizer uses	4.2	
Manure N production[9]	6.0	14
Human waste N[10]	1.3	3
Total *Terrestrial* inputs	**43.5**	**100**
Nr inputs to the *Aquatic* environmental system		
Surface water N flux[11]	4.8	

*Terms with an asterisk indicate Nr that is created, highlighting where reactive nitrogen is introduced to the environment.

Table 1 Notes

a. The Nr estimates in this table are shown with two significant digits or 0.1 million metric tons N per year (or Tg N/yr) to reflect their uncertainty; occasionally this report will show data to more significant digits, strictly for numerical accuracy. Because numbers are rounded, the sums of source category inputs do not always equal the total reported inputs. Obtaining quantitative estimates of each of the Nr terms and the associated uncertainties remains a major scientific challenge.

b. Reducing the uncertainty in total deposition of atmospheric Nr to the surface of the 48 contiguous United States remains a scientific and policy priority. Based on observations and models, we estimate 5.9 (range 4 - 9) Tg N/yr total anthropogenic Nr deposition to the entire 48 States. The EPA sponsored Community Multiscale Air Quality (CMAQ) Model run yielded a value of 4.8 Tg N/yr. The value shown for the total (6.9 Tg N/yr) reflects the assumption that organo-nitrogen species should be added to the model estimate as 30% of the total.

c. The synthetic N total includes 5.8 Tg N of fertilizer net imports to the United States (8.25 Tg N imported - 2.41 Tg N exported) plus 9.4 Tg N of fertilizer produced in the United States in 2002.

Table 1 Data Sources (all data reflect N fluxes in the United States in 2002)

[1] Emissions, N_2O-N (U.S. EPA Inventory of U.S. Greenhouse Gas Emissions and Sinks, 1990-2006)

[2] Emissions, NH_x-N; Emissions, NO_x-N (U.S. EPA National Emissions Inventory, release version October 2007)

[3] Atmospheric deposition, organic N (30% of total atmospheric N deposition, Neff et al. 2002)

[4] Atmospheric deposition, inorganic NO_y-N and NH_x-N (U.S. EPA CMAQ model)

[5] N_2 fixation in cultivated croplands (USDA census of agriculture 2002, literature coefficients)

[6] N_2 fixation in non-cultivated vegetation (unpublished data estimate after Cleveland and Asner, 1990)

[7] Net N imports in commodities and fertilizer trade (FAO - FAOSTAT)

[8] Synthetic N fertilizer use (FAO - FAOSTAT and Association of American Plant Food Control Officials - AAPFCO)

[9] Manure N production (USDA census of agriculture, literature coefficients)

[10] Human waste N (U.S. Census Bureau population census, literature coefficients)

[11] Surface water N flux (USGS SPARROW model, after Alexander et al., 2008)

2.2. Sources of New Nr to the Environment

Creation of "new" Nr in the environment refers to Nr that is either newly fixed within or transported into the United States. This "new" Nr highlights where Nr is introduced into ecosystems. New Nr arises from fossil fuel combustion, food production, and materials production (Table 1).

Fossil fuel combustion emits Nr (mostly NO_x) to the atmosphere. Fossil fuel combustion introduces 3.5 Tg N/yr and 1.9 Tg N/yr of NO_x-N to the atmosphere from transportation, and utility/other industry sources, respectively (Table 1). Another 0.2 Tg N/yr of NH_3-N and 0.1 Tg N/yr of N_2O-N is emitted from the same sources (Table 1). Thus the total amount of Nr created by fossil fuel combustion is 5.7 Tg N/yr, of which > 90% is in the form of NO_x-N.

Synthetic Nr fertilizers are typically produced by the Haber-Bosch process and used primarily in agriculture to support food production. Production of fertilizers within the U.S. introduces Nr into U.S. terrestrial landscapes at the rate of 9.4 Tg N/yr, and net imports of fertilizer via world trade introduce 5.8 Tg N/yr. Of this total (15.2 Tg N/yr), 9.8 Tg N/yr is used as fertilizer on farms and 1.1 Tg N/yr is used on non-farms (i.e., residential and recreational turf-grass and gardens, and in explosives used by the mining industry), and 4.2 Tg N/yr is introduced for non-fertilizer uses, such as for production of plastics, fibers, resins, and for additives to animal feed (Table 1).

Additional Nr is introduced into the U.S. from C-BNF by agricultural legume crops such as soybean and alfalfa (7.7 Tg N/yr), and from imports of N contained in grain and meat (0.15 Tg N/yr) (Table 1).

Thus in 2002, anthropogenic activities introduced a total of 29 Tg N into the U.S., mostly in support of food production, although turf production, industrial uses and fossil fuel combustion were also important sources. Natural sources of Nr in the U.S. are biological nitrogen fixation (BNF) in unmanaged landscapes, and lightning. The former contributes 6.4 Tg N/yr (Table 1) and the latter 0.1 Tg N/yr. Clearly, anthropogenic activities dominate the introduction of Nr into the U.S.

Losses of Nr to the environment in the U.S. occur during fossil fuel combustion and food production. The former occurs immediately, as Nr formation during combustion is inadvertent and the Nr, primarily as NO_x, is emitted directly into the atmosphere. The latter occurs through all stages of food production and consumption. The following four Sections (2.2.1-2.2.4) of this report document the magnitude of the losses of Nr to the environment from the various components of both energy and food production.

2.2.1. Nr Formation and Losses to the Environment from Fossil Fuel Combustion

Fossil fuels such as coal, petroleum, and natural gas provide about 80% of all energy production in the U.S. (based on year 2000). When these fuels are burned at high temperatures, NO_x is formed. The source of N is either the N contained in the fossil fuel or the N_2 that makes up about 80% of atmosphere. Fuel-derived N is important in the case of burning coal (which contains N), while atmospheric-derived N_2 is transformed to NO_x during higher temperature processes that occur when gasoline or diesel fuel is burned in motor vehicles (Table 1). As Figure 3 indicates, in the U.S., highway motor vehicles account for the largest anthropogenic source of NO_x (36%), followed by off-highway vehicles, electric utilities, and industrial processes. Emissions from aircraft make up only about 1% of the U.S. total for NO_x, but a large proportion of this is released in free troposphere where lifetimes are long and adverse impacts wide-ranging. As such, continued reductions are encouraged (e.g., EPA Regulatory Announcement: New Emission Standards for New Commercial Aircraft Engines, www.epa.gov/oms/regs/nonroad/aviation/420f05015.htm).

Figure 3 also illustrates that the amount of NO_x (reported as metric tons of N) released from various fossil fuel sources has decreased dramatically from 1970. Total emissions were on the order of 7,400 metric tons in 1970 and decreased to 5,900 in 2002, with further decreases in 2006 to 5,030 metric tons. Overall this represents a decrease of over 30%. The top sources (highway vehicles, off-highway vehicles, electric utilities, and other industrial and combustion systems) show decreases of 15-30% from 1990 to 2002 (Figure 4). Reductions were the highest for "other" systems followed by electric utilities. These decreases are most likely the result of changes in regulations and control technologies for these stationary systems. More recent preliminary information provided by EPA indicates that electric power NO_x emissions may have decreased 70% between 1990 and 2009 and that the electric power sector's NO_x emissions now account for about 12% of anthropogenic NO_x emissions in the U.S. (U.S. EPA, 2010b). To a lesser extent, changes in highway vehicle regulations and the removal of older fleets from the road has resulted in a decrease of approximately 15%. This decrease however, is accompanied by an increase in miles traveled, which suggests that the actual decrease in a single vehicle is larger. Off-highway vehicles showed an increase in emissions, potentially due to better quantification of these sources. Such sources include locomotives and marine engines. EPA is in the process of implementing a number of regulations that will reduce NO_x emission from mobile sources (see Appendix F). Figure 5 (provided by EPA) projects decreases in U.S. mobile-source NO_x emissions. The implications of these recent regulations are not reflected in the quantitative analyses presented in this report. However, additional control of these and other sources could further decrease emissions. In fact, technological development in the locomotive industry shows that decreases of approximately 70% are possible. Further decreases would require more innovative, expensive methods such as selective catalytic reduction

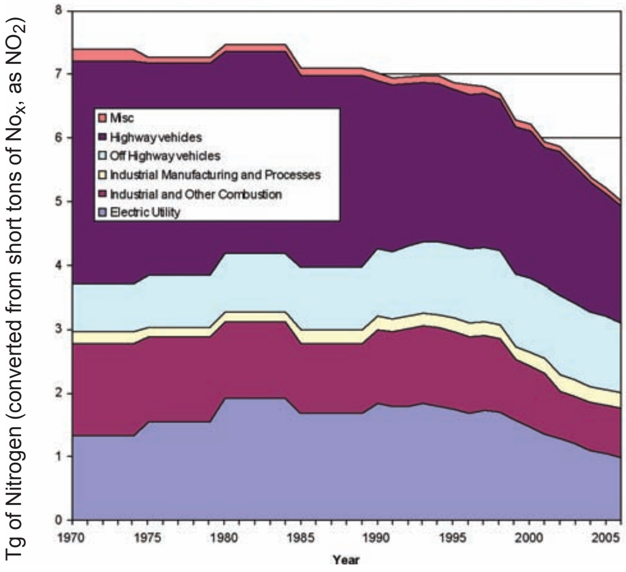

Figure 3: U.S. NO_x emission trends, 1970-2006

Data are reported as thousands of metric tons of N converted from NO_x as NO_2

Data source: www.epa.gov/ttn/chief/trends. More recent information provided by EPA indicates that electric power NO_x emissions decreased 70% between 1990 and 2009 (U.S. EPA, 2010b).

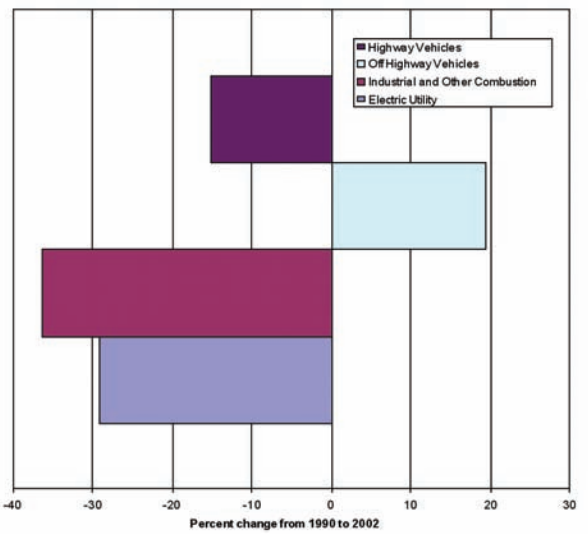

Figure 4: Percent reductions in NO_x emissions, 1990-2002, from different sources (off-road, on-road, power generation, etc.)

Data source: www.epa.gov/ttn/chief/trends. More recent preliminary information (U.S. Environmental Protection Agency, 2010b) indicates that electric power NO_x emissions may have decreased 70% between 1990 and 2009.

(SCR) with urea injection. Engine manufacturers are also investigating using SCR systems for diesels. However, it must be noted that these systems emit small amounts of NH_3 and must be operated properly to avoid trading off NO_x emissions for NH_3 emissions.

It should also be noted that it is difficult to control nitrogen emissions by regulating one source solely. As seen in Table 2 (data taken from 2001 for illustrative purposes have been reported with a higher degree of precision than is likely to be known), in Texas, fuel combustion sources are on the same order as emissions from highway vehicles. By comparison, in California, highway and off-highway vehicles are the dominant source of nitrogen emissions (over 75%). These results can be attributed to differences in the two states' industries and power plants. In Texas,almost 40% of the power generation is from coal-fired plants. On the other hand, California imports most of its coal-fired power and generates its own power predominantly from other sources, such as natural gas (50%), hydro and nuclear (33%). Table 2 also shows emissions in Florida, Ohio, and Illinois. The emission of NO_x from highway vehicles is likely related to population. For example, the estimated population of California in 2006 was 36.4 million people versus Ohio and Illinois which are on the order of 11-12 million.

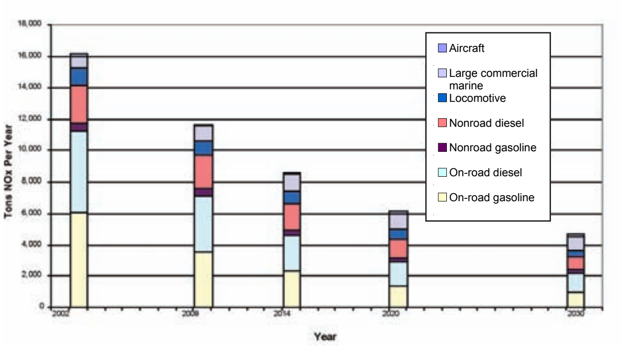

Figure 5: Mobile source NO_x emission inventories

Source: Figure provided by Margaret Zawacki of the U.S. EPA Office of Transportation and Air Quality. Inventory data used to develop this figure are available in EPA's final rule, Control of Emissions from New Marine Compression-Ignition Engines at or Above 30 Liters per Cylinder (www.epa.gov/otaq/regs/nonroad/marine/ci/420r09019.pdf), except for onroad emissions, which EPA generated by running MOVES2010 (www.epa.gov/otaq/models/moves/index.htm) at the national-month level.

2.2.2. Nr Inputs and Losses to the Environment from Crop Agriculture

Agriculture uses more Nr and accounts for more Nr losses to the environment than any other economic sector. Synthetic fertilizers are the largest sources of Nr input to agricultural systems. The next largest source is N fixation in cultivated croplands (Table 1). The major pathways by which Nr is lost from these systems include NO_3^- losses from leaching, runoff and erosion and gaseous emissions via volatilization of NH_3 and NO_x and nitrification/denitrification. Similar loss pathways occur for Nr that cycles through livestock systems, which also account for a large portion of Nr flux (predominantly as NH_3) in animal agricultural systems (Aneja et al., 2006). Therefore, assessment of Nr impacts on the environment and development of strategies to minimize negative impact should be based on a thorough understanding and accurate accounting

Table 2: Examples of multiple sources from states with high NO_x emissions based on 2001 data, and tons of NO_x as NO_2

	TX	CA	FL	OH	IL
Fuel Combustion – Electric Util.	91,441	8,441	87,489	93,792	59,124
Fuel Combustion – Industrial	98,978	31,237	11,792	17,300	26,481
Fuel Combustion – Other	9,222	21,407	5,707	12,974	10,894
Industrial Processes	25,584	13,786	5,933	8,123	7,122
Highway Vehicles	164,937	182,471	116,889	83,593	78,278
Off-Highway Vehicles	106,162	85,064	38,475	46,239	52,797
Miscellaneous Sources	4,807	7,882	13,110	1,526	999
TOTAL, metric tons	**501,151**	**350,301**	**279,778**	**263,561**	**235,817**

Source: Data derived from the 2001 information obtained at: www.epa.gov/air/data/.

of Nr fluxes in both crop and livestock systems, and the trends in management practices that have greatest influence on Nr loss to the environment from these systems (Aneja et al, 2008a,c).

As previously noted, in the past 60 years N fertilizers have had a beneficial effect on agriculture both nationally and globally by increasing crop yields. However, the high loading of Nr from agricultural nutrient sources has led to deleterious effects on the environment, such as decreased visibility from increased aerosol production and elevated N concentration in the atmosphere, ground, and surface waters (Galloway et al., 2003).

Nitrogen fertilizer use information

Obtaining accurate data on fertilizer use is a critical first step in understanding Nr cycles in agriculture. There are several sources of data reporting fertilizer usage but it is not clear whether data quality are sufficient for assessing environmental impact. Although the Uniform Fertilizer Tonnage Reporting System (UFTRS) was developed to collect fees to fund the consumer protection mission of state chemists and fertilizer regulatory control officials, it also provides data on fertilizer sales in many states, which in turn are used by many agencies and environmental scientists to estimate consumption and use of nitrogenous fertilizers in the U.S. The Association of American Plant Food Control Officials (AAPFCO) tallies and publishes the statewide fertilizer sales data annually (Terry and Kirby, 2006). Information published by AAPFCO is a widely used source of fertilizer use data. It is typically assumed that fertilizers are used in the same region in which they were sold. The annual data published by AAPFCO, which are based on commercial fertilizer sold (and taxed in some but not all states), is the only state-level data source available. This state-level data source includes fertilizer sales for both agricultural and non-agricultural purposes. These state-level data must then be allocated to counties, regions, or watersheds in the states, and the algorithms used for this process are based on a number of assumptions that address dealer/farmer storage, inventories, and cross-state sales issues (personal communication, Stan Daberkow, USDA-ERS).

The U.S. Department of Agriculture (USDA) National Agricultural Statistics Service (NASS) fertilizer usage data represents another source of information derived from farmer “agricultural chemical use” surveys that provide information in six categories: field crops, fruits and vegetables, nurseries/floriculture, livestock use, and post-harvest application. NASS periodically collects fertilizer, pesticide, and pest management data from a stratified random sample of farmers at the field level. The NASS report represents a useful data source but also would require extrapolation across reported crop acreage to represent a complete sample of application rates.

The UFTRS was not designed to track the source of inorganic nutrients applied to agricultural land on the geographic scale needed for watershed modeling. The system only tracks sales of synthetic fertilizers and not manure or biosolids applied to farmland. In addition, geographical data associated with each sale may or may not be near the actual point of application. However, given either regulatory or legislative changes (data reporting is mandated through each state’s fertilizer law), it could be possible to refine the current system used by each state’s department of agriculture to generate more precise data for improved modeling of watershed-scale nutrient mass balances. Those changes would help target interventions and extension programs to improve nutrient management and reduce nutrient losses. The lack of potential funding and the necessity to coordinate all the states involved limit the practicality of such an approach.

State departments of agriculture have considered ways to improve the reporting system. Such improvements could include:

1. An assessment to determine the needs for fertilizer usage data, the accuracy of the current data collection methods, and whether methods require revision to meet highest priority needs
2. Improvements in the database format and web-based access
3. The identification of funding sources to support development of a more accurate, accessible, and comprehensive database system
4. Education and outreach to improve precision of reported fertilizer tonnage, including a clear distinction between nutrients used in crop, livestock, and non-agricultural operations

In addition, the information could be refined to reflect site-specific data layers, although that would require development of a geospatial framework (and legal authority) to encourage reporting at the retail level where it is possible to collect geographic information.

The Chesapeake Bay watershed provides a good example of the fertilizer data dilemma. While the fertilizer tonnage that is currently being utilized to calibrate the Chesapeake Bay Program Watershed Model is relatively accurate, the county-specific tonnage may have an accuracy of only ±20 to 50% (Chesapeake Bay Scientific Technical Advisory Committee, 2007). For example, in a recent year, 17% of the reported tonnage was reported without an identified use, and there are indications some tonnage may have been reported more than once through the distribution chain (Chesapeake Bay Scientific Technical Advisory Committee, 2007). It is also possible that fertilizer reported for crop agriculture may actually have been used for lawn and turf, forestry, or other non-agriculture applications.

Nitrogen fertilizer application data collected on a specific crop-by-crop basis that can be associated with crop yields and location are essential for assessing both use patterns and efficiency. As previously mentioned, the USDA-NASS maintains a database on N fertilizer rates applied to the major crops (corn, wheat, cotton, soybeans, and occasionally other crops). The data are gathered from farmer surveys conducted every other year. These data represent a source of useful information (*Protocols for Farming Reporting*, Mark R. Miller, USDA, NASS).

Data derived from NASS farmer surveys include six categories: field crops, fruits and vegetables, nurseries/floriculture, livestock use, and post-harvest application. For each group, NASS collects fertilizer, pesticide, and pest management data on a stratified random sample of farmers at the field level. One field represents an entire farm for each sample in the field crops survey. Fruit and vegetable information are collected for the entire farm. If the field chosen for sampling has had manure applied in conjunction with inorganic fertilizer, only the inorganic portion will be reported because the survey does not ask about manure.

Certain core crops are surveyed every other year on an even/odd basis and states are selected to cover at least 80% of planted acres. There has been a recent five-year gap in the collection of NASS data for corn and other commodity crops. NASS was scheduled to resume coverage of corn and other commodity crops in 2010. This is a critical data gap and it is a problem given the large changes in corn price and production area during this period. Those data are needed to make progress in assessing fertilizer use and efficiency for major crops in the U.S. USDA NASS must resume their yearly data collection for commodity crops (Chemical Use Survey). Potential environmental impacts of increased N inputs associated with expanded corn acreage for biofuel production cannot be properly evaluated in the absence of such critical nutrient management data.

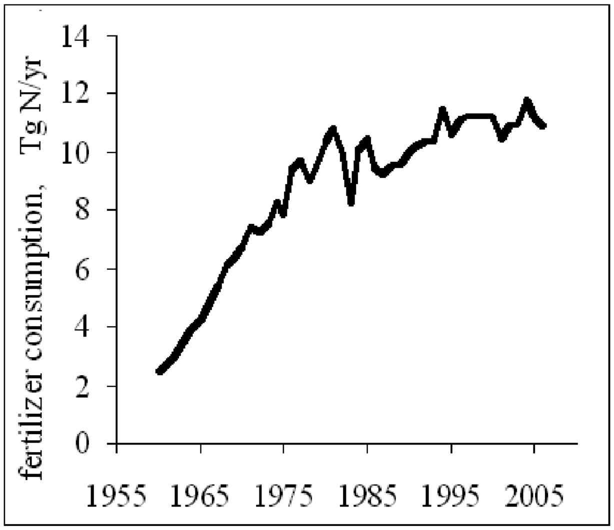

Figure 6: Fertilizer consumption in the United States, 1960 to 2006

Source: Slater et al., 2010. Reprinted with permission from the Association of American Plant Food Control Officials.

Based on the NASS survey data, the USDA Economic Research Service released a report on fertilizer use that provided data on fertilizer consumption and type of fertilizer used from 1960-2006 (Figure 6) and types of fertilizers used (Table 3) (USDA, 2008). The share of crop area receiving fertilizer and fertilizer use per receiving acre, by nutrient, are presented for the major producing states for corn, cotton, soybeans, and wheat. Additional data include fertilizer farm prices and indices of wholesale fertilizer price.

Finding 1: Crop agriculture receives 60% of U.S. annual new Nr inputs from anthropogenic sources (9.8 Tg from N fertilizer, 7.7 Tg from crop BNF versus 29 Tg total) and accounts for 58% (7.6 Tg) of total U.S. Nr losses from terrestrial systems to air and aquatic ecosystems. However, current monitoring of fertilizer use statistics by federal agencies is inadequate to accurately track trends in quantities and fate of N applied to major crops and the geospatial pattern by major watersheds.

Recommendation 1: *The Committee recommends increasing the specificity and regularity of data acquisition for fertilizer application to major agricultural crops in terms of timing and at a sufficiently small application scale (and also for urban residential and recreational turf) by county (or watershed) to better inform decision-making about policies and mitigation options for reducing Nr load in these systems, and to facilitate monitoring and evaluation of impact from implemented policies and mitigation efforts.*

Nitrogen fertilizer use efficiency

Nitrogen fertilizer use efficiency (NFUE) is critical because higher use efficiency leaves less N remaining to create potential environmental problems. Here and throughout this report we define NFUE as the grain yield per unit of applied N, which is the product of two parameters: (1) the proportion of applied N fertilizer that is taken up by the crop, or N fertilizer recovery efficiency [recovery efficiency (RE) in kg N uptake per kg N applied], and (2) the physiological efficiency with which the N taken up by the crop is used to produce economic yield for crops such as grain or fruit [physiological efficiency (PE) in kg yield per kg N uptake] (Cassman et al., 2002)[13]. All else equal, when higher NFUE is achieved without yield reduction, the crop takes up more of the applied N and incorporates it into its biomass, which leaves less of the applied Nr at risk for losses via leaching, volatilization, or denitrification. Fixen (2005) reports that there is substantial opportunity for increasing NFUE through development

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[13] N fertilizer use efficiency (NFUE) is calculated as the ratio of grain yield to the quantity of applied N fertilizer (kg grain/kg applied N).

Table 3: Types and amount of nitrogen fertilizers used in the United States in 2002

Synthetic Nitrogen Fertilizers	Tg N/year	% of total
Other	0.21	2
Urea	2.21	20
N Solutions	2.55	23
Anhydrous NH_3	2.88	26
Ammonium phosphates and N-P-K blends	2.28	21
Ammonium sulfate, aqua ammonia, ammonium nitrate, and other nitrate and ammonical N fertilizers	0.76	7
Total *	**10.89**	**100**

Data from Terry and Kirby, 2006

* Because of number rounding, the sum of individual percentages does not equal 100%.

and adoption of more sophisticated nutrient management decision aids.

In most cropping systems, RE is the most important determinant of NFUE. A recent review of RE for cereals based on field studies around the world, mostly conducted on “small-plot” experiments at research stations, reported mean single year RE values for maize, wheat and rice of 65%, 57%, and 46%, respectively (Ladha et al., 2005). However, crop RE values based on actual measurements in production-scale fields are seldom greater than 50% and often less than 33%. For example, a review of RE in different cropping systems estimated average recoveries of 37% for maize in the north central U.S. (Cassman et al., 2002). It is also important to note that soil N provides the majority of the N taken up by most crops grown on soils with moderate to good soil fertility. For maize in the U.S. Corn Belt, for example, 45-77% of total N uptake was estimated to come from soil N reserves, based on experiments from research stations (Sawyer et al., 2006). Therefore, highest N efficiency and economic return on N inputs are achieved when the amount and timing of applied N is synchronized with the availability of soil N throughout the growing season to minimize both the quantity of N input required and the N losses from soil and applied N sources.

However, there are relatively few data that provide direct measurement of N fertilizer recoveries by major field crops under production-scale conditions. Reducing the uncertainty in estimates of N fertilizer RE is fundamental for prioritization of research and education investments, both in the public and private sectors. While management can substantially improve RE on average, in any given year weather will always be an uncontrolled factor that can significantly influence system efficiency. Weather can influence system efficiency through effects on crop growth vigor and ability to acquire applied nutrients and through losses of nutrients due to runoff, denitrification, and leaching that can occur in periods of excessive rainfall.

Although total N fertilizer use in the U.S. has increased more slowly in the past two decades (Figure 6), yields of all major crops have continued to increase. Because crop yields are closely related to N uptake (Cassman et al., 2002), these trends imply a steady increase in NFUE and reduced N losses to the environment because more of the applied N is held in crop biomass and harvested grain. Greater NFUE has resulted from two factors. The first factor is a steady improvement in the stress tolerance of corn hybrids (Duvick and Cassman, 1999) that increases crop growth rates and allows sowing at higher plant densities, which together accelerate the establishment of a vigorous root system to intercept and acquire available N in the soil profile. The second factor is the development and adoption of technologies that may improve the congruence between crop N demand and the N supply for indigenous soil resources and applied N. Examples of such technologies include soil testing for residual nitrate and adjusting N fertilizer rates accordingly, split N fertilizer applications, fertigation (the application of nutrients through irrigation systems), site-specific management, and new fertilizer formulations (e.g., controlled release, nitrification inhibitors). For maize, which receives the largest share of total N fertilizer in the U.S. (44% in 2005), NFUE decreased markedly in the 1960s because N fertilizer rates rose more quickly than maize yields. However, with recognition of negative impact from over-application of N and associated N losses to the environment (especially with regard to water quality) investment in research and education to improve N fertilizer efficiency resulted in more than 50% increase in NFUE from 1974-76 to 2002-05 (Figure 7). Similar

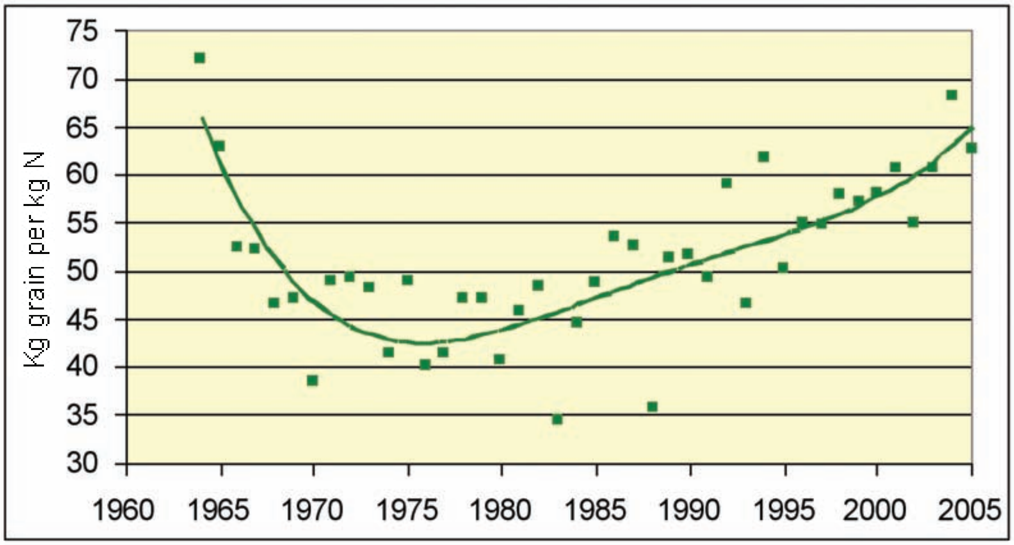

Figure 7: Trends in corn grain produced per unit of applied fertilizer N (NFUE) in the United States

Source: Adapted from Fixen and West, 2002 (Figure 6). Adapted with permission; Copyright 2002, Springer Science+Business Media B.V. on behalf of the Royal Swedish Academy of Sciences.

improvements have been documented for rice production in Japan and for overall crop production in Canada.

Despite these steady improvements, current levels of N fertilizer uptake efficiency appear to be relatively low, although data from production-scale studies are few (Cassman et al., 2002). Most farmers do not use best management practices (BMPs) with regard to nitrogen fertilizer management. For example, a recent U.S. Department of Agriculture Economic Research Service (USDA-ERS) Agricultural Resources and Environmental Indicators (AREI) report indicates that a majority of farmers still apply N in the fall, which gives the lowest fertilizer uptake efficiency and highest Nr losses compared to application in spring or during the crop growth period (USDA, ERS, 2006). This situation suggests substantial potential for improvement in NFUE and an associated reduction in Nr losses from crop agriculture, especially for maize in the warmer portions of the Corn Belt and other southern and southeast areas where maize is grown. One potential development is the use of controlled release fertilizers that release N in congruence with crop demand during the growing season. Although such fertilizers are already in use on high value horticultural crops, they are currently too expensive for lower value commodity grains such as corn, rice, or wheat. Control of N release should result in higher NFUE where there is high risk for N losses in cereal systems that receive the total amount of applied N in one or two large doses. Production-scale field studies are needed to document the benefits of this and other innovative technologies to improve NFUE.

As producers have significantly increased yields in commodity crops over the past 25 years, it is questionable whether university recommendations for nutrient applications are still current. Many university recommendations are now 20 to 25 years old. As a corollary to this problem, numerous environmental models of nutrient pollution are still utilizing older yield estimates, which often underestimate crop nutrient uptake and overestimate nutrient losses (Burgholzer, 2007).

A systematic effort needs to be made to update data on crop yields used to estimate nutrient losses. The concept of NFUE should be emphasized as a way to address the need to balance economic *and* environmental goals. In fact, the development and adoption of technologies that improve nitrogen fertilizer efficiency can contribute to more profitable cropping systems through a reduction in fertilizer costs. For example, average NFUE in the U.S. required 1.0 kg of applied N to produce 43 kg of grain yield in the 1974-1976 period, whereas that same amount of N produced 65 kg of grain in the 2003-2005 period (data taken from Figure 7). This gain in efficiency means that it is possible to achieve the 2004 U.S. average corn yield of about 150 bushels per acre (9,444 kg/ha) with 144 lbs per acre (161 kg/ha) of applied N fertilizer (based on the most recent NFUE achieved by U.S. corn producers) versus about 200 lbs per acre (224 kg/ha) of N fertilizer at the 1980 efficiency level.

Nitrogen costs have become extremely volatile, mirroring natural gas prices. In late 2008, N fertilizer prices were more than double the 2006-2007 N fertilizer prices. More recently, N fertilizer prices have fallen back to two thirds of the high following the decline of natural gas prices. If corn can be sold for $4.00 per bushel (25.5 kg) and N costs $0.40 a pound (0.45 kg), this is a 10 to 1 price ratio – the same as the $2.00 corn and $0.20 nitrogen ratio that was typical from 2000 to 2005. There are also other critical factors in a farmer’s N application decisions, such as yield at the margin and weather. In the Corn Belt, one or two years in five may provide extremely favorable weather for corn production. A producer may view applying some extra N, hoping for good weather, as a reasonable economic gamble. If the yield response is more than half a bushel (12.7 kg) of corn per pound (0.45 kg) of N at the margin or if there is more than one extremely good year in five, the farmer benefits.

Realistically, few farmers calculate their marginal returns from additional N in good years versus average, but the high corn-to-fertilizer price ratio encourages some farmers to plan for a good year and consider a larger N application than might otherwise be appropriate for the N utilization in the four years of lower yield. This presents a real dilemma if the policy goal is to reduce N transfers to the environment, especially in the four years of average or lower yields. Meeting this challenge will require approaches such as the development of real-time, in-season, decision-making tools that allow crop producers to use N fertilizer rates for average yields at planting and during early vegetative growth, and a final top-dressing as required to meet any additional N demand above this amount due to favorable climate and soil conditions that support higher than average yields (Cassman, 1999; Cassman et al., 2002). Robust crop simulation models using real-time climate data at a relatively localized geographic scale will be required to develop such tools.

Another option is to develop new, alternative crop production systems that require less N fertilizer. Such systems may employ legume cover crops, more diverse crop rotations, and tighter integration between crop and livestock production to achieve greater reliance on N inputs from legume N fixation and recycling of N in manure and compost. At issue, however, is whether such systems actually reduce Nr losses to the environment because the same loss mechanisms and pathways operate on N from both commercial fertilizer and organic sources. Also at issue is the indirect land use change impact from widespread adoption of these more diverse cropping systems because they have reduced crop yields per unit land area compared to more simplified crop rotations such as corn-soybeans that receive N fertilizer. Lower yields would require more land in production to meet food demand. Therefore, a key issue is whether the tradeoff in reduced N fertilizer inputs to more diverse crop rotations with organic N inputs would actually result in less Nr losses to the environment compared to conventional cropping systems that require less land to produce the same amount of crop output.

Another approach to reduce Nr losses from agriculture would be to shift and/or adjust cropping systems across the landscape. This would involve changes in land use as well as crops. This approach for parametric reductions in nitrogen was analyzed extensively in the Gulf of Mexico Hypoxia Assessment (Doering et al., 1999). As part of a modeling exercise to address opportunities and consequences for reducing Nr, crop rotations as well as tillage practices and fertilizer inputs were adjusted to meet successive constraints on excessive Nr while also maximizing consumer and producer welfare to the extent possible. The model favored those crops and cropping practices that had lower Nr leakage. Where the model could not find a crop production system at given locations that allowed positive net returns to the land, that land was taken out of production. At a 20 percent Nr reduction scenario, crop acreage was reduced by about 6%. This analysis was based on crop genetics, rotations, and tillage practices as of the 1990s.

Unintended impacts of lower application rates of nitrogen for crop production

Crop production and environmental quality are potentially lost or gained at the expense of each other. Although leakage of N from crop production systems cannot be eliminated, N losses can be minimized substantially. One way to decrease leakage is to apply less N fertilizer to croplands. For example, Hu et al. (2007), using the SWAT model, predict that decreasing N fertilizer application rates by 10 to 50% of those used in the 1990s in the upper Embrarras River watershed in east central Illinois would decrease NO_3^- output to the river by 10 to 43%. This simple “solution” can cause problems for crop production as yields and crop quality (protein content) may decrease, causing economic loss to the farmer, decreased food quality for the consumer and, at a global scale, a reduction in food security.

Cropping systems managed in a consistent manner over time reach a functional equilibrium between nitrogen inputs and outputs. Because crop yields are closely linked to the quantity of N accumulation in above ground biomass at maturity (Cassman et al., 2002), there would be a proportional decrease in crop yields in response to a decrease in the amount of N fertilizer application. The magnitude of this yield reduction would depend on the magnitude of decrease in the rate of applied N and the efficiency of N uptake from the applied N, as well as interrelationships with the availability of other nutrients. Hence, yield reductions can be mitigated, or even eliminated, if methods and fertilizer formulations used in fertilizer-N application increase the efficiency of N uptake to offset the reduction in the amount of applied N. It is also important to note that reduced or insufficient N rates for crop production risk impairment of long-term soil productivity. Jaynes and Karlen (2005) reported that N rates below the agronomic and economic optimum could degrade the soil resource and decrease soil organic matter over time. Thus care must be exercised in any N rate adjustments to protect soil productivity and to support soil resource sustainability.

Another concern associated with decreasing N fertilizer input to cereal crop production is the effect on crop quality. In Denmark, for example, national efforts to decrease Nr losses to the environment resulted in a large reduction in use of N fertilizer in crop production (Figure 8). This situation occurred as part of Denmark’s

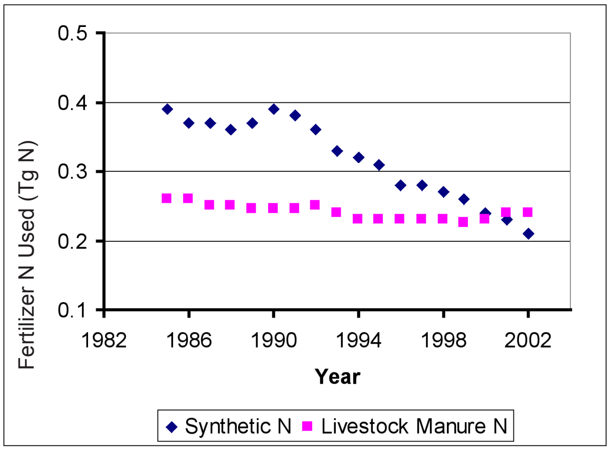

Figure 8: Synthetic fertilizer and livestock manure N used as fertilizer in Denmark 1985-2003

Source: IFA, 2004. Reprinted with permission from the International Fertilizer Industry Association.

response to the European Union Nitrate Directive. Synthetic fertilizer N use in Denmark decreased from approximately 0.4 Tg N in 1991 to 0.2 Tg N in 2002. Animal manure N application decreased from 0.25 Tg N to approximately 0.24 Tg N during this time period. Despite this large decrease in N fertilizer use, cereal crop yields remained relatively constant (data not shown). But while grain yields were maintained, there was a decrease in grain quality as determined by protein content (Figure 9). Grain protein content in wheat is critical for determining its quality for bread. In the U.S., a grain protein content of 12% is considered the threshold for good quality bread wheat, and N fertilizer application rate has a large influence on determining this trait (Cassman et al., 1992). As can be seen, grain protein content has declined from 12 to 10% in Denmark over the same period of lower fertilizer application rates.

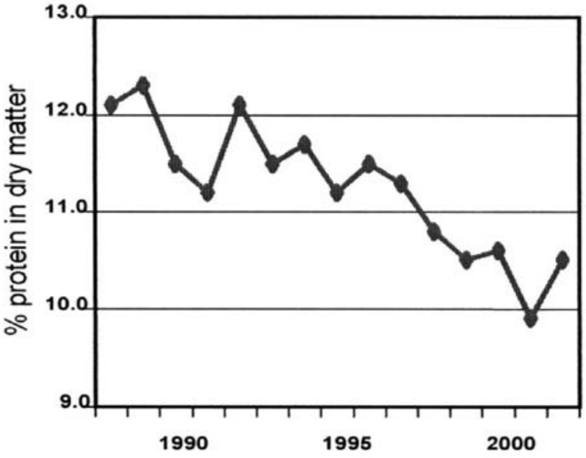

Figure 9: Protein content of cereal grain in Denmark

Source: IFA, 2004. Reprinted with permission from the International Fertilizer Industry Association.

Such trends raise several questions if declines continue or are found to be widespread. What is the cost to the farmer (considering human nutrition and end-use value costs)? Do these costs offset the environmental benefits created by decreasing N flows from crop production areas? What would be the regional and global impact if similar reductions in nitrogen fertilizer inputs to agriculture were put in place in developed countries that represent the largest source of grain exports to international markets?

As previously discussed, it is possible to increase crop yields without an increase in N fertilizer input if the methods used to apply N are modified to improve NFUE as illustrated by U.S. maize yield trends and fertilizer use (Figure 7). However, at some point continued yield increases will require additional N input because of the tight relationship between yield and crop N uptake requirements. The goal of N fertilizer management should therefore be to achieve highest possible NFUE while also sustaining crop yield increases adequate to meet food demand without need for a large expansion of crop production area. In fact, studies that evaluate NFUE in production agriculture find a wide range in NFUE among farmers. For farmers who achieve high NFUE, a reduction in N fertilizer rate would reduce yields. Hence, mandating an across-the-board reduction in N fertilizer use penalizes the good farmers who achieve high NFUE as their yields would likely fall while it would have little effect on yields of farmers who currently over fertilize (Cassman et al., 2003; Dobermann and Cassman, 2004).

Finally, if yields were significantly reduced as a result of lower N fertilization rates, more land may need to be brought into production. Because nearly all prime agricultural land is already used for crop production, expansion of crop area will most likely occur on more marginal land, such as the land currently in the Conservation Reserve Program (CRP). Such conversion would have two negative results. First, additional N losses from these acres would occur due to relatively low N fertilizer efficiency that typically occurs on marginal land that has multiple soil constraints to crop growth and yield. Second, conversion from CRP to crop production would result in loss of soil organic matter that accumulates under grassland, which in turn would lead to increased greenhouse gas emissions from agriculture.

Finding 2: Nr inputs to crop systems are critical to sustain crop productivity and soil quality. Moreover, given limited land and water resources, global population growth, and rapid economic development in the world's most populous countries, the challenge is to accelerate increases in crop yields on existing farm land while also achieving a substantial increase in N fertilizer uptake efficiency. This process is called "ecological intensification" because it recognizes the need to meet future food, feed, fiber, and energy demand of a growing human population while also protecting environmental quality and ecosystem services for future generations (Cassman, 1999). More diverse cropping systems with decreased Nr fertilizer input may also provide an option to increase NFUE on a large scale if the decrease in Nr losses per unit of crop production in these diverse systems can be achieved without a decrease in total food production, which would trigger indirect land use change to replace the lost production and negate the benefits. However, crop cultivars and agronomic practices are changing rapidly, which changes N requirements, but current efforts in research, extension, and conservation programs on N management within these rapidly evolving systems are not adequate to meet the challenge of providing better information to increase NFUE.

Recommendation 2: *To obtain better information on Nr inputs and crop productivity, the Committee recommends that:*

Recommendation 2a: *Data on NFUE and N mass balance, based on direct measurements from production-scale fields, should be generated for the major crops to identify which cropping systems and regions are of greatest concern with regard to mitigation of Nr load and to better focus research investments, policy development, and prioritization of risk mitigation strategies.*

Recommendation 2b: *Efforts at USDA and universities should be promoted to: (i) investigate means to increase the rate of gain in crop yields on existing farm land while increasing N fertilizer uptake efficiency and (ii) explore the potential for more diverse cropping systems with lower N fertilizer input requirements so long as large-scale adoption of such systems would not cause indirect land use change.*

Recommendation 2c: *EPA should work closely with the USDA, Department of Energy, and the National Science Foundation, and universities to help identify research and education priorities to support more efficient use and better mitigation of Nr applied to agricultural systems.*

Biological fixation in cultivated croplands

Reactive nitrogen is also introduced to the landscape in significant quantities via BNF in cultivated crop lands. Management of biologically fixed N, insofar as it is possible, is proportionally as critical a task as the management of synthetic N because Nr from BNF is prone to the same loss pathways as Nr from commercial fertilizers. To quantify BNF due to human cultivation of crops, the Committee calculated the annual agricultural fixation for 2002 using crop areas and yields reported by the USDA Census of Agriculture (USDA, 2002). The Committee multiplied the area planted in leguminous crop species by the rate of N fixation specific to each crop type, assigning rates based on a literature review, as summarized in Table 4 below and shown relative to other inputs in Table 1. Annual nitrogen inputs to cropping system from BNF by legume crops was 7.7 Tg N/yr in 2002, accounting for ~15% of the overall Nr inputs to the terrestrial landscape from all sources and 20% of the agricultural sources (Table 1). Soybean and alfalfa contributions are the most important agricultural legumes in terms of nitrogen input and contribute 69% of total BNF inputs in U.S. agriculture.

Emissions factors and losses to the environment from fertilizers and organic nitrogen sources

Agriculture is a significant contributor of Nr inputs into the atmosphere. Nitrogen fertilizer losses vary greatly

Table 4: Estimates of nitrogen input from biological nitrogen fixation from major legume crops, hay, and pasture

Nr fixation in cultivated croplands				
	production area, Mha	rate, kg/ha/yr	Tg N/yr	% of total *
Soybeans	29.3	111	3.25	42
Alfalfa	9.16	224	2.05	27
Other leguminous hay	15.4	117	1.80	23
Western pasture	161	1	0.16	2
Eastern pasture	22.0	15	0.33	4
Dry beans, peas, lentils	0.88	90	0.08	1
Total			**7.67**	**100**

Source: Updated estimate for soybean based on a generalized relationship between soybean yield and the quantity of N fixation (Salvagiotti et al., 2008). Other values are from Boyer et al. (2002).

* Because of number rounding, the sum of individual percentages does not equal 100%.

due to differences in soil properties, climate, and the method, form, amount, timing and placement of applied N (Cassman et al., 2002). In addition, any factor that affects crop growth vigor and root system function also affects the ability of the plant to recover applied N efficiently. For example, denitrification can range from 0% to 70% of applied N (Aulakh et al., 1992). This process is mediated by heterotrophic, facultative anaerobic soil bacteria that are most active under warm, wet soil conditions; they have low activity in dry sandy soils.

Despite this variation, watershed, regional, and national assessments of carbon and N cycling often rely on average values for losses from each pathway. For example, the Intergovernmental Panel on Climate Change (IPCC) assumes that 1% of applied N fertilizer (uncertainty range of 0.3-3.0%) is lost from direct emissions of N_2O at the field level due to nitrification/denitrification. This assumption is based on analysis of all appropriate scientific publications that report these losses for specific crops and cropping systems (IPCC, 2007a). The same 1% default emission factor for field-level N_2O emission is applied to other N inputs from crop residues, organic amendments such as manure, and from mineralization of native soil organic matter. Data from scores of field studies were used to obtain this average value. A number of recent studies confirm that N_2O losses to the environment during the growing season at the field level can represent <1% of the applied nitrogen – even in intensive, high-yield cropping systems (Adviento-Borbe et al., 2006). Despite these average values, it is also clear that N_2O losses can vary widely even within the same field and from year to year due to normal variation in climate and crop management (Parkin and Kaspar, 2006; Snyder et al., 2007). Moreover, the loss of nitrogen from agricultural watersheds is strongly dependent on climate change (e.g., rainfall changes). Predicted increases and decreases in rainfall will likely have a dramatic impact on nitrogen export from agricultural fields. For example, precipitation is predicted to increase in the upper Mississippi watershed and, other factors being equal, N export should increase (e.g., Justic et al., 1995a,b).

Additional indirect N_2O emissions result from denitrification of volatilized NH_3 deposited elsewhere or from NO_3^- lost to leaching and runoff as the Nr cascades through other ecosystems after leaving the field to which it was applied. Here the IPCC assessment protocol assumes that volatilization losses represent 10% of applied N, and that N_2O emissions for these losses are 1% of this amount; leaching losses are assumed to be 30% of applied nitrogen and N_2O emissions are 0.75% of that amount (IPCC, 2007a). Therefore, the IPCC default value for total direct and indirect N_2O emissions represents about 1.4% of the applied N from fertilizer. By the same calculations, 1.4% of the N in applied organic matter, either as manure or compost or in recycled crop residues, is also assumed to be emitted as N_2O. Recent work funded by EPA used the DAYCENT model to estimate N_2O emissions from cropping systems (Del Grosso et al., 2005). However, due to the cost of field validation of such models, there are relatively few validations across a representative range of cropping systems and environment. Therefore, it is not clear that use of such a complex model gives better estimates of N_2O emissions than the more straightforward IPCC assessment protocol.

Others have estimated higher average global N_2O losses of 3-5% of applied nitrogen fertilizer based on historical changes in atmospheric N_2O content and changes in Nr production during the past 50 years (Crutzen et al., 2008), as opposed to the field-based estimates that form the basis of IPCC estimates. Because N_2O is such a potent GHG, and given the more than two-fold difference in estimates of N_2O losses, there is a critical need to improve understanding and prediction of N_2O losses from agricultural systems. N_2O emissions in the U.S. are estimated to be 0.78 Tg N/yr (Table 5) (U.S. EPA, 2005b).

Table 5: N_2O emissions in the United States, 2002

	Tg N/yr	% *
Agricultural Soil Management	0.54	69
Manure Management	0.03	4
Mobile Combustion	0.09	12
Stationary Combustion	0.03	4
Nitric & Adipic Acid Production	0.05	6
Wastewater Treatment	0.02	2
Other	0.02	2
Total	**0.78**	**100**

* Because of number rounding, the sum of individual percentages does not equal 100%.

Biogenic NO_x emissions from croplands are on the order of 0.5% of fertilizer input – much more than this in sandy soils and less as clay content increases (Aneja et al. 1996; Sullivan et al. 1996; Veldkamp and Keller, 1997; Civerolo and Dickerson, 1998). However, NO_x emissions by agricultural burning are relatively unimportant. Ammonia volatilization of N from applied fertilizer can be the dominant pathway of N loss in rice soils and can account for 0% to more than 50% of the applied N depending on water management, soil properties, and method of application (citations within Peoples et al., 1995, 2004). Ammonia volatilization can be of the same range in upland cropping systems, with largest losses occurring typically on alkaline soils (Peoples et al., 1995, 2004). The IPCC (2007a) uses a value of 10% of synthetic fertilizer N application and 20% of manure N as estimates of average NH_3 volatilization.

Taken together, N losses from all forms of direct gaseous emissions from crop production systems can represent a substantial portion of applied N fertilizer when soil conditions favor such emissions and there is a lack of synchrony between the amount of N applied and the immediate crop demand (Goulding, 2004). Therefore, achieving greater congruence between crop demand and the N supply from fertilizer is a key management tactic to reduce N losses from all sources. Success in reducing N losses and emissions from agriculture will depend on increased efforts in research and extension to close gaps in our understanding of N cycling and management in crop production, especially as systems further intensify to meet rapidly expanding demand for food, feed, fiber, and biofuel.

Finding 3: Nitrous oxide emissions from the Nr inputs to cropland from fertilizer, manure, and legume fixation represent a large proportion of agriculture's contribution to GHG emissions, and the importance of this source of anthropogenic GHG will likely increase unless NFUE is markedly improved in crop production systems. Despite its importance, there is considerable uncertainty in the estimates of nitrous oxide emissions from fertilizer and research should focus on reducing this uncertainty.

Recommendation 3: *The Committee recommends that EPA ensure that the uncertainty in estimates of nitrous oxide emissions from crop agriculture be greatly reduced through the conduct of EPA research and through coordination of research efforts more generally with other agencies such as USDA, DOE, NSF and with research conducted at universities.*

Impact of biofuel production capacity on Nr flux in agriculture

The enormous use of liquid fuels in the U.S., the rising demand for petroleum based liquid fuels from countries like China and India, and the decline in petroleum discovery all contributed to the recent record high petroleum prices. In addition, most of the world's petroleum reserves are located in politically unstable areas. This has provided strong motivation for policies promoting investment in biofuels made from corn, oil crops, and ultimately from cellulosic materials. In the U.S., ethanol production capacity from corn more than doubled from 2006 to 2009 (to a capacity of over 47 billion liters/year in January 2009). The renewable fuels standard in the 2007 Energy Independence and Security Act (EISA) will support another 9.5 billion liters/year of corn-based ethanol by 2015. An additional 79.5 billion liters is to come from cellulosic ethanol by 2022. Production of biodiesel from vegetable oils also is encouraged in EISA, but expansion has been slowed by the high food value of such oils. Brazil is rapidly expanding its production of relatively low cost sugarcane ethanol and U.S. policies continue to be aimed at bringing about increased future biofuel production in the U.S.

In 2007 and 2008 petroleum prices pushed ethanol prices high enough to draw corn from food and feed uses into ethanol production and contribute to the increased price of corn. Because of the increase in petroleum/ethanol prices and the government subsidy for ethanol production, 30% of the corn crop was used to produce ethanol in 2008 (Abbott et. al., 2008). With the subsequent collapse in petroleum and ethanol prices, followed by corn prices, there has been unused capacity in the U.S. ethanol industry as the corn/ethanol price ratio made ethanol production uneconomic for some firms. However, EISA is likely to lead to the production of cellulosic materials and even some expanded corn production for biofuels once the U.S. gets beyond the current blending limit for ethanol (Doering and Tyner, 2009). The higher corn prices of 2007 and 2008 resulted in more land being planted to corn and higher N fertilizer requirements. Corn area went from 78.41 million acres (31.73 million hectares) in 2006-2007 to 93.6 million acres (37.88 million hectares) in 2007-2008. Reduction in soybean production accounted for 12.01 million acres (4.86 million hectares) of the corn expansion, and the remaining new acreage came primarily from reduced cotton acreage and from hayland and pasture. This strong response to high demand for biofuel feedstock has led to concern about increased pressure on the environment from biofuels. One important factor is the increased N necessary for growing corn and cellulosic materials (Robertson, et. al. 2008). Biofuels production and consumption will also result in NO_x emissions to the atmosphere. Expansion of corn or cellulosic materials production into marginal lands can be even more problematic with respect to nutrient leaching and soil erosion. Changes in N fertilizer prices add uncertainty to the additional amounts of N that may ultimately be used in biofuel feedstock production. Production of large amounts of distillers grains co-product is also changing the way that livestock feed rations are formulated, which in turn could have an influence on the cycling of N in cattle manure (Klopfenstein et al., 2008).

In February 2010, EPA released its final rule for the expanded Renewable Fuels Standard (RFS2) regarding greenhouse gas emissions from various types of biofuels based on life-cycle analysis as required by the 2007 EISA. Standards for corn ethanol and soybean biodiesel are based on studies that include data from large-scale production systems because these are the only biofuels currently produced on a large commercial scale. In contrast, life cycle analysis (LCA) standards for cellulosic and other advanced biofuels were based on data from "pilot- and bench-scale" studies, or in many cases on hypotheses and rough estimates. While these initial estimates meet requirements imposed by 2007 EISA, the science underpinning life cycle assessments of biofuel systems, including direct and indirect land use change, are relatively undeveloped and evolving rapidly.

Finding 4: Rapid expansion of biofuel production has the potential to increase N fertilizer use through expanding corn production and its associated N fertilizer inputs and extending cultivation for cellulosic materials that will also need N. Distillers grains are changing animal diets and affecting N recycling in livestock. Both have important consequences for effective future management of Nr, but current models and understanding of how expanded biofuel production will affect Nr inputs and outputs from agriculture are inadequate to guide policy.

Recommendation 4: *EPA should work with USDA and universities to improve understanding and prediction of how expansion of biofuel production, as mandated by the 2007 EISA, will affect Nr inputs and outputs from agriculture and livestock systems. Rapid expansion of biofuel production has the potential to increase N fertilizer use through expansion of corn production area and associated N fertilizer inputs, and from extending cultivation of cellulosic materials that will also need N inputs. Current models and understanding are not adequate to guide policy on how to minimize impact of biofuel expansion on environmental concerns related to Nr.*

2.2.3. Nr Inputs and Losses from Animal Agriculture

In the U.S., domestic animals produce 6.0 Tg N/yr in manure and are the largest source of atmospheric NH_3-N (1.6 Tg N/yr) (Table 1). Livestock also contribute to N_2O-N emissions, though in much smaller proportions (~4% of total U.S. N_2O-N emissions).

Trends in Animal Agriculture

While animal production has been increasing since World War II, this report emphasizes the period from 1970 to 2006. The production of chicken broilers increased more than four fold from 1970 to 2006 (Figure 10) and milk production increased by nearly 60% in this time period (Figure 11). Turkey production doubled and pork production increased about 25%, while meat from cattle (beef and dairy) remained constant (Figure 10).

Another trend in animal production has been for fewer animals to produce more animal products. For example, the 60% increase in the amount of milk produced in 2006 compared to 1970 required 25% fewer cows (Figures 11 and 12). Animal inventories declined by 10% for beef brood cows from 36 million head in 1970 to 33 million head in 2006, and the inventory of breeder pigs and market hogs declined 8% from 673 million head to 625 million head in the same period, even with similar or greater annual meat production. This trend resulted from

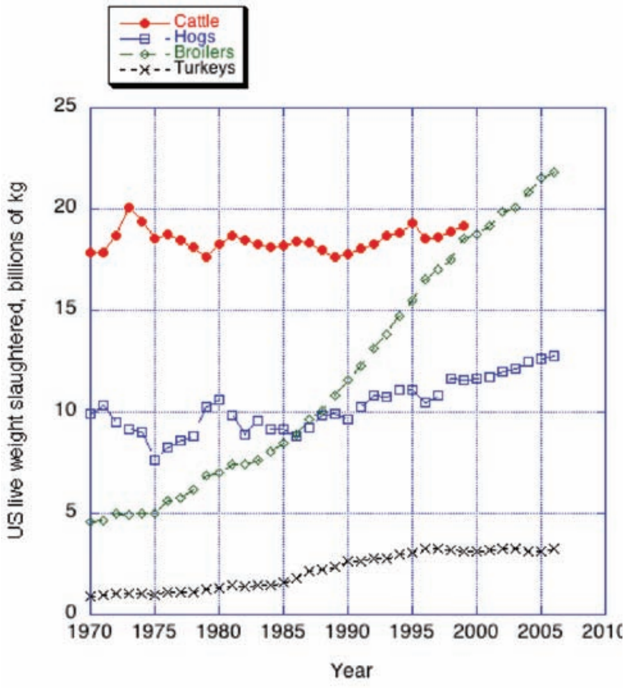

Figure 10: Meat production from 1970 to 2006

Source: USDA, NASS, 2007 – Census Reports. Data on cattle were not taken after 1999.

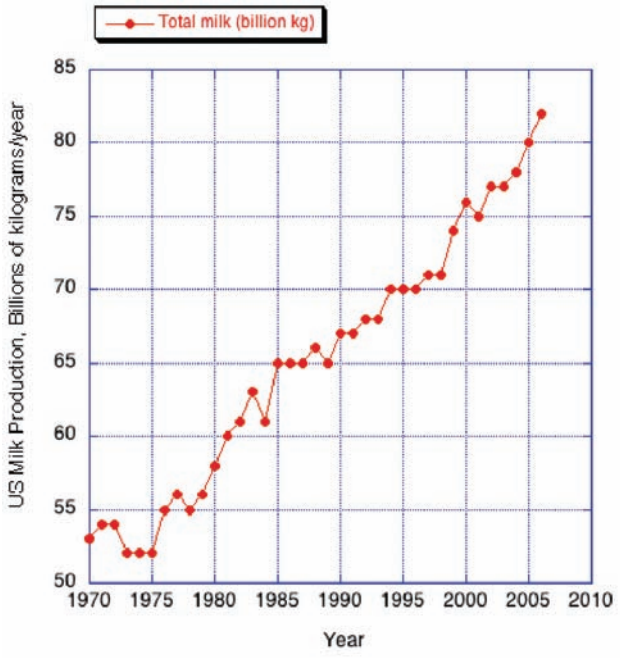

Figure 11: Milk production from 1970 to 2006

Source: USDA, NASS, 2007 – Census Reports greater growth rates of animals producing more meat in a shorter amount of time. In 1970, broilers were slaughtered after 80 days on feed at 1.7 kg live weight, but by 2006 the average weight was 2.5 kg after only 44 days on feed (USDA, NASS, 2007).

Another trend in animal agriculture has been the increased size and smaller number of animal operations, which results from the mechanization of agricultural practices and increased specialization. There were only 7% as many swine operations and 11% as many dairy operations in 2006 as there were in 1970 (Figure 13). There were half as many beef operations in 2006 as in 1970, but beef operations also expanded in size while smaller producers held jobs off the farm.

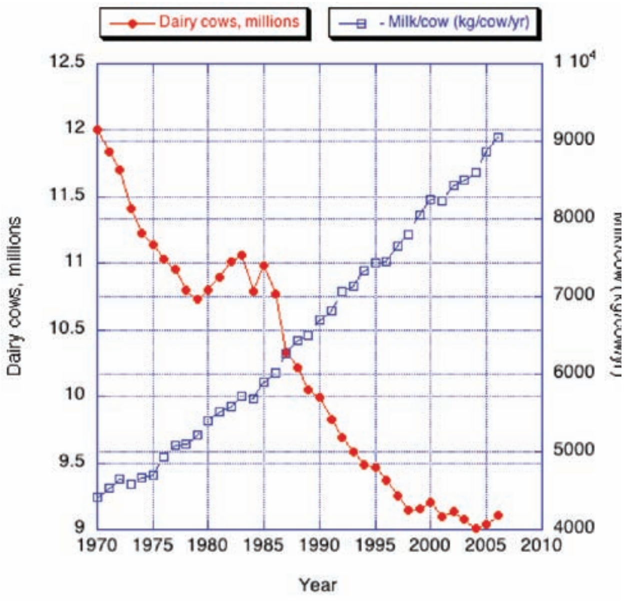

Figure 12: U.S. inventory of mature dairy cows and milk production per cow from 1970 to 2006

Source: USDA, NASS, 2007 – Census Reports

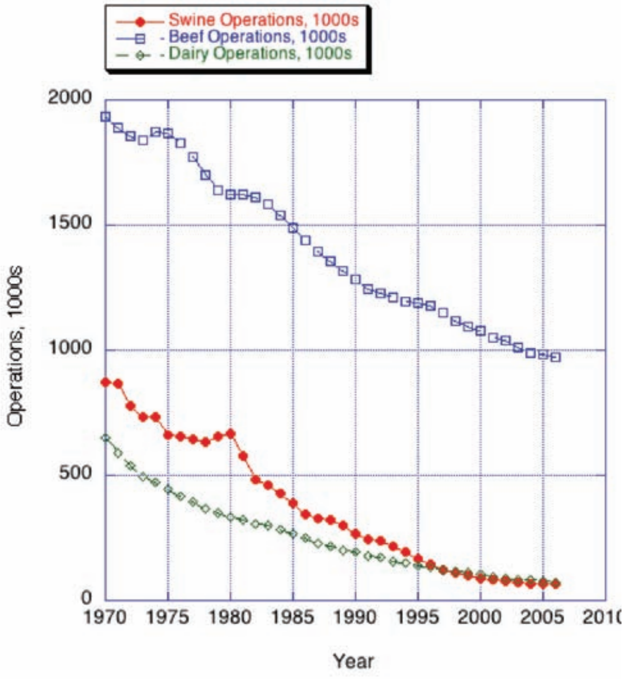

Figure 13: Number of animal operations in the United States from 1970 to 2006

Source: USDA, NASS, 2007 – Census Reports

All of these trends show an increase in management and labor efficiency to produce a similar or greater amount of animal products. Also, because animal production is more concentrated on fewer farms with greater specialization, fewer crops are produced on those farms. As a result, it is increasingly common to have more manure nutrients produced on a livestock farm than can be used efficiently as fertilizer for crops on that farm. Therefore, unless the manure is applied over a larger crop area, the resulting over-application of manure on the livestock farm can reduce the subsequent efficiency of its utilization and result in greater nutrient losses.

Impact of livestock production trends on nitrogen use efficiency

The trends in livestock production have both positive and negative environmental impacts. One of the significant positive impacts is that with smaller animal inventories producing greater quantities of animal products, there is an improved efficiency of nitrogen utilization per product produced. This effect is partly the result of effectively reducing maintenance requirements during production. The requirements for feeding animals can be divided into two components: maintenance and production. The maintenance component is the feed that is used to keep the animal alive and healthy so that production is possible. The production component includes the feed that is converted to animal protein and waste due to the inefficiencies of these conversions. The maintenance component depends upon the number of animals, each animal's mass, and the time the animal is on feed. Thus, the maintenance requirement is diluted by faster growth rates and greater body weight at slaughter. The increases in production rates over time have led to greater efficiencies in N and P utilization for animal production and lower amounts of nutrients excreted per unit of animal protein produced.

Public concerns about the potential environmental and health effect of air emissions from CAFOs expand the impacts of food production beyond those associated with traditional agricultural practices (NRC, 2001; Aneja et al., 2009). Increased emissions of N compounds from animal agriculture into the atmosphere may lead to increased odor and interactions in atmospheric reactions (e.g., gas-to-particle conversion) (Baek et al., 2004a; Baek and Aneja, 2004b). These compounds are then transported by wind and returned to the surface by wet and dry deposition processes, which may have adverse effects on human health and the environment (McMurry et. al, 2004; Aneja et. al, 2006b, 2008a,b,c; Galloway et. al, 2008).

Adverse effects, further discussed in Chapter 3 of this report, include eutrophication, soil acidification, loss of biodiversity, and reactions that increase the mass concentration of atmospheric aerosols ($PM_{2.5}$). Aerosol formation occurs when HNO_3 reacts with basic compounds, and NH_3 reacts with compounds. Ecosystem acidification can occur when HNO_3 is deposited from the atmosphere. In addition, acidification can also occur when NH_x is deposited due to the production of HNO_3 from nitrification via soil microbes. Soil acidification occurs when HNO_3 or NH_4^+ deposits on soils with low buffering capacity and this can cause growth limitations to sensitive plant species. Deposition of NO_3^- or NH_4^+ also causes eutrophication (i.e., an over-abundance of nutrients), which can promote harmful algal growth leading to the decline of aquatic species. In fact, volatized NO_3^- can travel hundreds of miles from its source, affecting local and regional biodiversity far from its origin (Aneja et al. 2008b; James, 2008).

The potential for reduced environmental impact from Nr in livestock systems depends on the proportion of the total intake attributable to maintenance costs. The commonly used tables for diet formulation published periodically by the NRC for various animal commodities can be used to track diet formulation practices and assumptions regarding maintenance and production requirements. About one third of the energy intake recommended for growing broilers was assumed to be needed for maintenance (NRC, 1994) but protein requirements were not divided between maintenance and production. For example, a dairy cow producing 40 kg milk per year would divert about 25% of its energy and 12% of its protein to maintenance (NRC, 2001).

In terms of nutritional efficiency of a herd or flock, maintenance of a productive phase (e.g., growth, lactation) also requires maintenance of a reproductive phase of the animal's life cycle. In other words, the actual nutritional maintenance cost of a herd or flock is greater than it is for productive individuals only. For example, milk production requires non-lactating cows and heifers in the herd which do not produce milk but which consume nutrients. These additional maintenance costs are lower for broiler flocks than for cattle.

Finding 5: There are no nationwide monitoring networks in the United States to quantify agricultural emissions of greenhouse gases, NO, N_2O, reduced sulfur compounds, volatile organic compounds (VOCs), and NH_3. Satellite observations of agricultural emissions hold promise for providing a high degree of spatial coverage and may complement surface observations in this network, although robust methods are yet to be developed. In contrast there is a large network in place to assess the changes in the chemical climate of the United States associated with fossil fuel energy production, i.e., the National Atmospheric Deposition Program/National Trends Network (NADP/NTN), which has been monitoring the wet deposition of sulfate (SO_4^{2-}), NO_3^-, and NH_4^+ since 1978.

Recommendation 5: *The status and trends of gases and particulate matter precursors emitted from agricultural emissions, e.g.,* NO_3^- *and* NH_4^+ *should be monitored and assessed utilizing a nationwide network of monitoring stations. EPA should coordinate and inform its regulatory monitoring and management of reactive nitrogen with the multiple efforts of all agencies including those of the U.S. Department of Agriculture and NSF supported efforts such as the National Ecological Observatory Network (NEON) and the Long Term Ecological Research Network (LTER).*

Changes in feeding practices

From 1970 to 2006, several feeding practices were changed for diets fed to livestock (NRC, 1977, 1988, 1996, 2001). In 1989 and 1996, the NRC introduced the idea of dividing the form of protein fed to ruminants into that which is degraded by rumen microorganisms and that which passes through the rumen to be digested directly in the stomach and small intestine. Feeding ruminants with attention to rumen degraded and rumen undegraded protein decreases the amount of protein fed by 10 to 15% for a given protein requirement. For poultry and swine, manufactured amino acids were added to diets, decreasing the need for protein by 30%. Today, two amino acids (lysine and methionine), coated in a way to prevent degradation in the rumen, are sometimes added to dairy cattle diets thereby decreasing protein intake by another 15% (NRC, 2001). Phytase added to swine and poultry diets in the past decade has decreased phosphorus feeding by 20 to 50% with some of the decrease attributed simply to better understanding phosphorus requirements.

It is difficult to estimate the combined effects of these changes in feeding practices, but we have calculated changes in manure N by using USDA NASS data and assuming improvements in both production rates and ration formulation (Table 6). In the case of beef cattle diet formulation, the changes in feeding practices were determined by comparing the NRC 1976 recommendations with the NRC 1996 recommendations. Surprisingly, NRC 1996 recommended greater total crude protein compared to NRC 1976 despite formulating for rumen degraded and un-degraded protein and considering amino acid content. Therefore, improved diet formulation did not decrease N intake for beef in this time range but the effect of reduced maintenance did improve efficiency of N utilization.

Reduced nitrogen excretion from increased efficiency

Nitrogen excretion as fraction of animal production decreased from 1970 to 2006 (Table 6). However, in cases where the total amount of animal production in the U.S. increased substantially (e.g., broilers), total N excretion increased. The decrease in N excretion per unit of animal

Table 6: Livestock N excretion per kg production (g/kg) and per total United States (Tg/yr)

Commodity*	1970		2006	
	g/kg product	Total United States	g/kg product	Total United States
Milk	17	0.89	11	0.92
Pork, live weight	57	0.56	42	0.54
Broilers, live weight	56	0.26	46	1.00
Beef, live weight	123	1.2	110	1.3

*Does not include manure produced for reproduction of stock (e.g., growing dairy heifers, breeder pigs).

productivity was estimated by calculating the effects of changes in feeding practices and reduction of maintenance as described previously. The data in Table 6 indicate that there has been an increase in N utilization efficiency for livestock products.

For broilers, data are available to more accurately estimate the effect of changes in feeding and genetics on N excretion over time. However, these data do not represent the time period of interest in this report. Havenstein et al. (1994) compared a 1957 strain of broiler fed a 1957 diet to a 1991 strain fed a 1991 diet. Based on the reported N intake and production data, there was a 51% reduction in N excreted between these diets (Kohn, 2004).

Similarly, Kohn (2004) compared N excreted by U.S. dairy cows in 1944 and 2001. In 1944, the historically largest herd of dairy cattle in the U.S. (25 million cows) produced an average of 7 kg milk per cow per day (USDA NASS, 2007). In 2001, nine million cows produced an average of 27 kg milk per cow per day. Assuming the cows in 1944 and 2001 were fed according to popular feeding recommendations of the time, the N intakes were 360 and 490 g/day per cow, and N excretion rates (N intake minus N in milk) were 326 and 364 g/day per cow. Multiplying by the number of cows in the U.S., shows that total milk production increased 40% from 52 billion kg to 73 billion kg, while N excretion decreased 60% from 3.0 Tg N to 1.2 Tg N, respectively.

Table 7 provides information on manure production from animal husbandry in the U.S. For Table 7, manure N was calculated for all U.S. animal agriculture using data on animal production from the 2002 Census of Agriculture (USDA, 2002). For data on livestock production (cattle, calves, poultry, hogs, and pigs), manure was calculated by the methods of Moffit and Lander (1999), following the exact methods they had used to compute manure from the 1997 Census of Agriculture, but using the updated information from the 2002 Census of Agriculture. For data on production of manure from other animals (horses, goats, and sheep), the table uses coefficients for manure excretion as a function of average animal weights and animal inventory, taken from Battye et al. (1994).

Table 7: Manure production from animal husbandry in the continental United States, Tg N per year 2002.

	Tg N/yr	%
Cattle & Calves	4.35	72
Poultry	0.94	16
Hogs & Pigs	0.53	9
Horses, Goats & Sheep	0.19	3
Continental United States*	6.02	100

* Because of number rounding, the sum of manure production by category does not equal the reported total for the continental U.S.

Volatilization of animal waste

Ammonia volatilization is highly variable and is influenced by the amount of total ammonical nitrogen (TAN), temperature, wind speed, pH, chemical and microbiological activities, diffusive and convective transport in the manure, and gas phase resistance in the boundary layer above the source (Arogo et al., 2006).

EPA estimates annual manure N excreted in livestock production in the U.S. for the *Inventory of U.S. Greenhouse Gas Emissions and Sinks* (U.S. EPA, 2007e). For the year 2002, these estimates indicate that a total of 6.8 Tg of N was excreted in livestock manure. Only a fraction of this N (~1.24 Tg) was recovered and applied directly as a nutrient source for crop production. Approximately 1.8 Tg N was transferred from the manure management systems, most likely by ammonia volatilization. Other loss vectors include leaching and runoff during treatment, and storage and transport before soil application. The remainder of the N was deposited in pastures and rangeland or in paddocks. This N is also susceptible to movement into the atmosphere and aquatic systems or incorporation into soil organic matter. By a combination of BMPs and engineered solutions it may be possible to reduce the emissions and discharge of odors, pathogens, and nitrogen compounds from agricultural operations (Aneja et al., 2008b,d).

Total manure production reported in Table 7 in the contiguous U.S. was estimated using USDA's method and yields an estimate of 6.0 Tg N/yr; while EPA's GHG inventory method in Table 8 yields a total for the U.S. of 6.8 Tg N/yr in 2002. The "greenhouse gas" method suggests 13% higher manure N production. This difference highlights uncertainty in the calculations. Although the values in Table 8 include Alaska and Hawaii, the values in Table 7 do not; however, given the relatively small amount of livestock production in those states, that does not contribute substantially to the difference.

Finding 6: Farm-level improvements in manure management can substantially reduce Nr load and transfer. While the NPDES permitting process for CAFOs does include nutrient and manure management plans to limit the transport of land applied nutrients off farms, substantial off-farm transport of Nr still occurs, especially via atmospheric transport. There are currently very few incentives or regulations to decrease these transfers and loads despite the existence of management options to mitigate.

Recommendation 6: *A policy, regulatory, and incentive framework is needed and should be developed to improve manure management to reduce Nr load and ammonia transfer, taking into account phosphorus load issues.*

2.2.4. Nr Inputs to Residential and Recreational Turf Systems

Turf grasses cover 12.6-16.2 million ha across the continental U.S. (Milesi et al., 2005). The area under turf grass is roughly the size of the New England states and occupies an area up to three times larger than that of *irrigated* corn (The Lawn Institute, 2007). The majority of this turf area (approximately 75%) is in residential lawns. About 80% of all U.S. households have private lawns (Templeton et al. 1998) that average 0.08 ha in size (Vinlove and Torla, 1995). Another approximately15 % of total turf grass area is in low-maintenance parks and approximately 10% is in athletic fields and golf courses, which often receive higher levels of N application due to hard use conditions.

Supplemental N fertilization is often necessary to maintain healthy and aesthetically pleasing turf color, high shoot density, and the ability to resist and recover from stress and damage. Nitrogen also may be derived from atmospheric deposition or recycled decomposition of soil and grass clipping organic matter. Whether these inputs are sufficient to maintain lawns of adequate quality depends on many factors including age of the turf, uses, and expectations or goals of the homeowner or field manager. Also, turf grasses are used to stabilize soil, often with an erosion-prevention matrix such as organic mats or with hydroseeding. Depending on circumstances, these turf uses may be temporary until natural vegetation succeeds the turf, or may be low-maintenance turfs that are seldom fertilized, such as highway medians and shoulders, grassy swales and buffers.

Turf grass is maintained under a variety of conditions. Approximately 50% of all turf grass is not fertilized; the remainder is fertilized at varied intensities (A.M. Petrovic, personal communication, June 5, 2007). The Committee has arrayed the different turf managements into the following three groups according to the estimated amount of N-fertilizer applied annually (Table 9): residential lawns maintained by homeowners (0.73 kg/100 m^2), residential lawns cared for by professional lawn care companies (2.92 [range, 1.95-7.3] kg/100 m^2), and athletic fields and golf courses (3.89 [range, 2.64-6.64] kg/100 m^2). The estimate of total N-fertilizer used

Table 8: Fate of livestock manure nitrogen (Tg N)

Activity	1990	1992	1994	1996	1998	2000	2002	2004
Managed manure N applied to major crops	1.1	1.2	1. 2	1.2	1.2	1.3	1.2	1.3
Manure N transferred from management systems	1.5	1.6	1.6	1.6	1.7	1.7	1.8	1.7
Pasture, range, & paddock manure N	3.9	4.0	4.1	4.2	3.9	3.8	3.8	3.7
Total *	**6.6**	**6.7**	**6.9**	**7.0**	**6.9**	**6.8**	**6.8**	**6.7**

Source: U.S. EPA, 2007e

* Because of number rounding, the sums of manure production do not always equal the reported totals.

Table 9: Estimate of fertilizer N used on turf grass in the United States in the year 2000, based on a total area of 12.6 million ha

Type of Turf Fertilized	Area (Million ha)	N rate (kg/ha/yr)*	Total N Used (Tg/yr)
Nominal Fertilization	4.7	73	0.35
Professional Lawn Care	0.93	296 (195-488)	0.27
High Maintenance Areas (golf/sports)	1.26	390	0.49
Total	**6.89**	–	**1.11**

Source: Data derived from Milesi; et al., 2005 and A.M. Petronic, Personal Communication 2007

*A conversion factor of 1000 m^2/ha was used with application rates (kg N/100 m^2) of: 0.73 for nominal fertilization, 2.92 for professional lawn care, and 3.89 for high maintenance areas kg N/100 m^2

on turf grass in the U.S. is 1.1 Tg N/year, or 9% of the total average annual N-fertilizer used between 1999 and 2005. Depending on land use patterns, certain areas of the country, particularly coastal areas where residential and urban properties prevail, turf fertilizer can be an important or even dominant source of nitrogen to surface waters.

Turf fertilizer N is susceptible to losses to the atmosphere and surface and ground water when it is not properly managed. Research on lawns has shown that leaching of NO_3^- can range between 0 and 50% of N applied (Petrovic, 1990). Nitrogen leaching losses can be greatly decreased by irrigating lightly and frequently, using multiple and light applications of fertilizers, fertilizing at the appropriate times (especially not too late in the growing season), and using soil tests to ensure proper balance of non-N soil condition and pH. In a soil column experiment with turf coverage, the percentage of N leached (as percentage of N applied) varied from 8% to 14% using light irrigation and from 2% to 37% with heavy irrigation. Applying fertilizer in appropriate amounts, avoiding periods when grass is dormant, and not fertilizing too soon before irrigation or large rainfall events can all help ensure leaching and runoff will be minimal without affecting turfgrass color and growth (Mangiafico and Guillard, 2006).

Nitrogen runoff losses are poorly quantified but a range similar to leaching is probable (A.M. Petrovic, personal communication). The chemical form of fertilizer N does not impact leaching/runoff unless the fertilizer is applied in late autumn (Petrovic, 2004a), although use of slow release or organic fertilizers can help reduce runoff and leaching. Shuman (2002) notes that runoff can be limited by applying minimum amounts of irrigation following fertilizer application and avoiding application before intense rain or when soil is wet. Losses of Nr to the atmosphere can be significant when urea is applied. Measured denitrification losses are usually small, but depend upon timing of N application relative to soil water status, irrigation, and temperature. Typically 25% of N applied is not accounted for in runoff, leaching, and uptake/removal, or soil sequestration (A.M. Petrovic, personal communication). This suggests that volatilization and denitrification are important loss vectors. Nitrogen volatilization (Beard and Kenna, 2008) rates range from 0.9% under light irrigation to 2.3% under heavy irrigation.

While under-fertilization can lead to reduced grass stand and weed encroachment which results in more leaching and runoff N losses than from well managed lawns (Petrovic and Larsson-Kovach, 1996; Petrovic, 2004b), Guillard (2008) recommends not fertilizing lawns of acceptable appearance. Further, prudent fertilization practices may include using one-third to one-half (or less) of the recommended application rate (i.e., application rates below 0.5 kg/100m2) and monitoring response (Guillard, 2008). Less or no fertilizer may produce acceptable lawns, especially once the lawn has matured, provided clippings are returned and mowing length is left high.

As noted above, according to Petrovic (personal communication) half the lawns in the U.S. may not receive any fertilizer. Those lawns are presumably satisfactory to their owners. Further N reductions can be made if white clover is incorporated into turf, and grasses such as fescues (which require little or no N supplements once mature) are selected for amenable parts of the country. These practices can potentially reduce N fertilization (and subsequent leaching risk) on turf by one third or more, saving 0.4 or more Tg N/year. When properly managed, turf grass provides a variety of services that include decreasing runoff, sequestering carbon dioxide, and providing a comfortable environment in which to live (Beard and Green, 1994).

Finding 7: Synthetic N fertilizer application to urban gardens and lawns amounts to approximately 9% of the total annual synthetic N fertilizer used in the United States. Even though this N represents a substantial portion of total N fertilizer use, the efficiency with which it is used receives relatively little attention.

Recommendation 7a: *To ensure that urban fertilizer is used as efficiently as possible, the Committee* *recommends that EPA work with other agencies such as USDA as well as state and local extension organizations to coordinate research to ensure that fertilization recommendations are accurate and promote awareness of the issue.*

Recommendation 7b: *Through outreach and education, supported by research, improved turf management practices should be promoted, including improved fertilizer application and formulation technologies and maintenance techniques that minimize supplemental Nr needs and losses, use of alternative turf varieties that require less fertilization, alternative ground covers in place of turf, and use of naturalistic landscaping that focuses on native species.*

2.3. Nr Transfer and Transformations in and Between Environmental Systems

This section discusses the transfers and flows of Nr within and between environmental systems (ES), which include atmospheric, terrestrial, and aquatic environments. The first Section (2.3.1) contains information on Nr deposition from the atmosphere to terrestrial and aquatic systems, presents estimates of input and recycling of Nr within terrestrial systems, and discusses movement of Nr from the terrestrial to the aquatic system. The second Section (2.3.2) presents an estimate of storage of Nr within the terrestrial system. Areas of uncertainty in Nr transfer and transformation are discussed in Section 2.3.3. Section 2.3.3 also contains an example analysis of Nr input and fate in 16 watersheds in the northeast U.S. No comprehensive national data are available to assess the transfer and transformations of Nr in and between the atmosphere, terrestrial systems and aquatic systems. The example analysis in Section 2.3.3 shows how an evaluation of inputs and fate of Nr could be conducted for a large watershed.

2.3.1. Input and Transfers of Nr in the United States

Nitrogen deposition from the atmosphere to the earth's surface

Atmospheric input contributes substantially to the Nr content of terrestrial and aquatic ecosystems for the U.S., but the magnitude and mechanisms of Nr deposition to the earth's surface remain major unanswered environmental questions. Along the eastern U.S. coast and eastern Gulf of Mexico, atmospheric deposition of N currently accounts for 10% to over 40% of new N loading to estuaries (Paerl et al., 2002). Other watershed contribution estimates range widely throughout the U.S., depending on size of the watershed, the size of the estuary, and the magnitude of contributing sources of atmospheric N enrichment. Valigura et al. (2001) identified a median atmospheric N contribution of about 15% for 42 watersheds located throughout the U.S., although the maximum estimate was 60%.

NO_x, NH_3 and their reaction products not deposited onto continents are generally lofted into the free troposphere where they can have a wide range of influence and, in the case of NO_x, because of nonlinearities in the photochemistry, generate substantial amounts of tropospheric ozone (U.S. EPA, 2006a). Total N deposition involves both gases and particles, and both dry and wet (in precipitation) processes. Rates of deposition for a given species (in units of mass of N per unit area per unit time) can be measured directly, inferred from mass balance of the atmospheric budget, or modeled numerically, but substantial uncertainties remain with each of these techniques when applied to deposition of any Nr species. A portion of the Nr deposited to the earth's surface is re-emitted as NH_3, NO, or N_2O (Galbally and Roy, 1978; Kim et al., 1994; Civerolo and Dickerson, 1998; IPCC, 2007a; Crutzen et al., 2008). Although naturally-produced Nr is involved, anthropogenic Nr dominates over most of the U.S. In Appendix A we provide a review of the state of the science concerning the total annual Nr deposition and trends in that deposition to the contiguous 48 states.

Deposition involves both oxidized and reduced N species. Of the oxidized forms of atmospheric N, all the members of the NO_y family (NO, NO_2, NO_3, N_2O_5, HONO, HNO_3, NO_3^-, PAN and other organo-nitrates, $RONO_2$) can be transferred from the troposphere to the surface, and some (e.g., NO) undergo bidirectional flux. Volatile amines are also detected as NO_y compounds (Kashihira et al., 1982; Wyers et al., 1993). Although a potent GHG, N_2O is only emitted, not deposited and therefore has not been considered in the material presented here and in Appendix A. Of the reduced forms of atmospheric nitrogen, NH_3 and NH_4^+ play a major role. There is also evidence of deposition of organic N such as amino acids and isoprene nitrates, and recent observations suggest that these can account for ~10% (possibly as much as 30%) of the U.S. NO_x budget, especially in summer (Keene et al., 2002; Horowitz et al., 2007; Duce et al., 2008; Sommariva et al., 2008). While this is a worthy research topic, measurements are still limited and deposition of organic N compounds has not been reviewed in this report. The wide array of relevant atmospheric compounds makes direct measurement and accurate load quantification challenging. Appendix A provides a discussion of wet and dry deposition of Nr in the U.S. and the relationship between emissions of Nr and observed deposition.

Consideration of NO_y as a supplement for the current NO_2 National Ambient Air Quality Standard

The six principal (or criteria) pollutants for which EPA has established National Ambient Air Quality Standards (NAAQS) include "oxides of nitrogen" (the sum of NO and NO_2) or NO_x. The specific chemical compound nitrogen dioxide (NO_2) has been selected as the indicator for compliance with the NAAQS for NO_x. The levels of primary and secondary standards for NO_2 are identical at 0.053 ppm (approximately 100 μg/m^3) in annual arithmetic average, calculated from the one-hour NO_2 concentrations. In forming an integrated policy for protecting the environment from adverse effects of reactive nitrogen, it is appropriate to consider whether the existing criteria pollutants are sufficiently inclusive of Nr species. EPA's recent Integrated Science Assessment for Oxides of Nitrogen and Sulfur – Ecological Criteria (U.S. EPA, 2008d) evaluated the scientific foundation for the review of the secondary (welfare-based) NAAQS for oxides of nitrogen and concluded that:

> *The instrumentation deployed at present in the routine monitoring networks for determination of gas-phase NO_2 and SO_2 concentrations is likely adequate for determining compliance with the current NAAQS. But in application for determining environmental effects, all these methods have important limitations, which make them inadequate for fully characterizing the state of the atmosphere at present, correctly representing the complex heterogeneity of N and S deposition across the landscape, and for realistically apportioning the contributions of reduced and oxidized forms of atmospheric N and S in driving observed biological effects at a national scale.*

Although the current standard for NO_2 is inadequate to protect welfare, there is a straightforward technical fix to this problem. NO_x ($NO + NO_2$) is a variable, often small component of reactive oxidized nitrogen (NO_y) as has been noted in EPA's Integrated Science Assessment for Oxides of Nitrogen and Sulfur – Ecological Criteria and extensively documented in the reviewed literature (Fahey et al., 1986; Fehsenfeld et al., 1987; Doddridge et al., 1991; Parrish et al., 1993; Poulida et al., 1994; Ridley et al., 1994; Emmons et al., 1997; Liang et al., 1998; Munger et al., 1998; Zhou et al., 2002; Takegawa et al., 2003; Parrish et al., 2004a,b; Horii et al., 2004, 2006; Dunlea et al., 2007; Kleinman et al., 2007; Hargrove and Zhang, 2008; Luria et al., 2008; Zhang et al., 2008; Schwab et al., 2009; Luke et al., 2010). The standard chemiluminescence NO/NO_x technique measures NO directly via reaction with O_3, and NO_x ($NO + NO_2$) by conversion of NO_2 to NO on hot molybdenum. It had been thought that NO_x would make up nearly all of NO_y in heavily polluted urban areas, but results including some from Mexico City (Dunlea et al., 2007) indicate that this is not the case. Interferences were up to 50% of the ambient NO_2 concentration even in a heavily urban area. Recent results from Houston, TX (Luke et al., 2010) showed a median NO_x/NO_y ratio of ~0.63 between 1300 and 1500 local time, a time of rapid ozone production. NO_2 has been used historically as the indicator for many health studies, and it is regulated in part because of studies that directly link it to respiratory effects. However, given the inadequacy of NO_2 as a standard to protect welfare, the Committee recommends that EPA reexamine the criteria pollutant oxides of nitrogen and the indicator species NO_2 and consider adding chemically reactive nitrogen as a criteria pollutant, and NH_x and NO_y as indicators to supplement the NO_2 National Ambient Air Quality Standard.

The references listed above also document that current suite of NO_x monitors are useful for proving compliance with the current NO_2 standard, but suffer substantial interferences and the extent of these interferences varies with time and location. Reactive N compounds, especially HNO_3, can be lost on the inlet components. Because the inlet does not transmit all reactive nitrogen compounds with high efficiency, commercial NO_x monitors may provide an upper limit for NO_x and a lower limit for NO_y, but they measure neither NO_x nor NO_y precisely. Numerical simulations do not currently produce consistent results for the partitioning of NO_y species, for example see Archibald et al., (2010). The data from the current monitoring network are of limited value for determining exposure to NO_2, for evaluating chemical transport models (CTMs), or for assessing efficacy of emissions control strategies (McClenny et al., 2002).

NO_y monitors differ from the NO_x monitors currently in use in the position of the hot molybdenum NO_2 to NO converter and in calibration and operation (e.g., Thermo Scientific Model 42i-Y). In an NO_x monitor the converter is well downstream of the inlet, behind a filter that removes particulate matter, while in an NO_y monitor the converter is at the inlet (Fehsenfeld et al., 1987; Doddridge et al., 1991; Doddridge et al., 1992; Parrish et al., 1993; Poulida et al., 1994; Ridley et al., 1994; Emmons et al., 1997; Liang et al., 1998; Munger et al., 1998; Parrish et al., 2004b; Dunlea et al., 2007; Hargrove and Zhang, 2008; Luria et al., 2008; Schwab et al., 2009). Thus the current monitors could be replaced or retrofitted to measure NO_y with a detection limit of 0.1 ppb at reasonable expense for equipment and for training operators. NO_y monitors, like NO_x monitors, provide proof of compliance with NO_2 standards. For specific measurement of NO_2, commercial instruments for selective NO_2 reduction (e.g., Air Quality Design, Inc., Wheat Ridge, CO) or direct NO_2 measurement are beginning to be available (Parrish and Fehsenfeld, 2000; Castellanos et al., 2009) and these may offer an alternative approach. Direct monitoring of NO_2 and other Nr species is a long-term goal, but new techniques will require thorough testing in polluted environments. NO_y monitors using hot molybdenum compare favorably with other NO_y techniques such as gold-catalyzed CO reduction and several studies have concluded that NO_y can be reliably measured in suburban and urban environments (Fehsenfeld et al., 1987; Crosley, 1996; Williams et al., 1998).

Conclusions on atmospheric deposition of Nr

As discussed here and in Appendix A, downward transport from the atmosphere is a major source of Nr to the earth's surface, but there are uncertainties in the characteristics and absolute magnitude of the flux. Pollutants not deposited are exported from the continent and alter the composition and radiative balance of the atmosphere on a large scale. A review of the literature revealed the following major points concerning the present state of the science:

1. Measurements from the National Atmospheric Deposition Program (NADP) indicate that wet deposition of ammonium plus nitrate for the period 2000-2006 averaged 3.1 kg N/ha/yr over the 48 contiguous states.
2. The reduced (NH_4^+) and oxidized (NO_3^-) forms of reactive N contributed about equally to the flux, but input to the eastern United States was greater (and less uncertain) than to the western U.S.
3. For the U.S. east of the Mississippi River, dry deposition data have also been analyzed – the Clean Air Standards and Trends Network (CASTNET) monitors vapor phase HNO_3, as well as particulate NO_3^- and NH_4^+. These measurements indicate 7.75 kg N/ha/yr total deposition (5.46 wet 2.29 dry) over the East. Conspicuous by its absence from this number is dry deposition of ammonia.
4. Decreases in NO_x emissions appear to have led to decreases in NO_3^- deposition. NADP data show a national decreasing trend in the wet nitrate deposition and some individual sites show statistically significant decreases in deposition and correlations with emissions.
5. A thorough review of all published studies of the U.S. NO_y budget indicates that about 70 % of the NO_x emitted by the U.S. is deposited onto the continent with the remainder exported, although substantial uncertainty remains. Major sources of uncertainty concerning the deposition of NO_y include dry deposition of unmonitored members of the NO_y family, uncertainties in the chemistry of organic N, and poorly constrained estimates of convective venting of the planetary boundary layer of the atmosphere (i.e., uncertainties concerning the venting of NO_y from the lowest layer of the atmosphere).
6. Based on observations and model estimates of the relative deposition of unmeasured quantities, total estimated deposition of all forms of Nr for the period 2000-2004 is ~11 kg N /ha /yr for the eastern U.S., and for the 48 states ~7.5 kg N /ha /yr with a range of 5.5 to 9.5 kg N /ha /yr.

Finding 8: Scientific uncertainty about the origins, transport, chemistry, sinks, and export of Nr remains high, but evidence is strong that atmospheric deposition of Nr to the earth's surface as well as emissions from the surface to the atmosphere contribute substantially to environmental and health problems. Nitrogen dioxide, NO_2, is often a small component of NO_y, the total of oxidized nitrogen in the atmosphere. The current NAAQS for NO_2, as an indicator of the criteria pollutant "oxides of nitrogen," is inadequate to protect health and welfare. NO_y should be considered seriously as a supplement or replacement for the NO_2 standard and in monitoring. Atmospheric emissions and concentrations of Nr from agricultural practices (primarily in the form of NH_3) have not been well monitored, but NH_4^+ ion concentration and wet deposition (as determined by NADP and NTN) appear to be increasing, suggesting that NH_3 emissions are increasing. Both wet and dry deposition contribute substantially to NH_x removal from the atmosphere, but only wet deposition is known with much scientific certainty. Thus consideration should be given to adding these chemically reduced and organic forms of Nr to the list of Criteria Pollutants.

Recommendation 8a: *EPA should reexamine the criteria pollutant "oxides of nitrogen" and the indicator species NO_2 and consider adding chemically reactive nitrogen as a criteria pollutant, and NH_x and NO_y as indicators to supplement the NO_2 National Ambient Air Quality Standard.*

Recommendation 8b: *Monitoring of NH_x and NO_y should begin as soon as possible to supplement the existing network of NO_2 compliance monitors.*

Recommendation 8c: *EPA should pursue the longer-term goal of monitoring individual components of Nr, such as NO_2 (with specificity), NO and PAN, and HNO_3, and other inorganic and reduced forms, as well as support the development of new measurement and monitoring methods.*

Recommendation 8d: *The scope and spatial coverage of the Nr concentration and flux monitoring networks (such as the National Atmospheric Deposition Program and the Clean Air Status and Trends Network) should be increased and an oversight review panel for these two networks should be appointed.*

Recommendation 8e: *EPA in coordination with other federal agencies should pursue research goals including:*

- *Measurements of deposition directly both at the CASTNET sites and in nearby locations with non-uniform surfaces such as forest edges.*
- *Improved measurements and models of convective venting of the planetary boundary layer and of long range transport.*
- *Improved analytical techniques and observations of atmospheric organic N compounds in vapor, particulate, and aqueous phases.*
- *Increased quality and spatial coverage of measurements of the NH_3 flux to the atmosphere from major sources especially agricultural practices.*
- *Improved measurement techniques for, and numerical models of NO_y and NH_x species (especially with regard to chemical transformations, surface deposition and offshore export, and linked ocean-land-atmosphere models of Nr).*

Input and recycling of Nr within terrestrial systems in the United States

Annual input of newly created Nr onto terrestrial ecosystems comes primarily from atmospheric deposition, synthetic fertilizer and BNF in managed and unmanaged ecosystems (Table 1). Although Nr deposited from the atmosphere to terrestrial systems is formed inadvertently during fossil fuel combustion and from volatilization of NH_3 from agricultural activities, it serves to provide nutrients, along with biological N fixation and synthetic fertilizer, for food, feed, and fiber production in the agricultural sector. Forests and grasslands use Nr for growth. Home gardens, parks and recreational areas utilize Nr within the urban landscape. Approximately 32 Tg of new Nr reached the land of the 48 contiguous states in 2002 (Table 1). An additional ~0.2 Tg of N was imported mainly as food and drink products (FAO, 2007). An additional ~12 Tg of Nr was recycled back to terrestrial and aquatic systems in livestock excreta (~6 Tg N), human excreta (~2 Tg N), and crop residue from the previous year's production (~4 Tg N; U.S. EPA, 2007e). Of this N, ~ 1.3 Tg (~1.2 from livestock manure and <0.1 from sewage sludge) was used as fertilizer for crop production (U.S. EPA, 2007e). More detailed information describing sources and cycling of reactive nitrogen input in terrestrial systems is provided in Appendix B.

Finding 9: Total N budgets within all terrestrial systems are highly uncertain. The relative amount of N losses from terrestrial systems ascribed to leaching, runoff, and denitrification are as uncertain as the N budgets themselves.

Recommendation 9: *EPA should join with USDA, DOE, and universities to work together in efforts to ensure that the N budgets of terrestrial systems are properly quantified and that the magnitudes of at least the major loss vectors are known.*

Transfer of Nr to aquatic systems

Within the nitrogen cascade, Nr flows from the atmosphere and terrestrial systems into aquatic systems. Aquatic systems include groundwater, wetlands, streams and rivers, lakes and the coastal marine environment. Nr is deposited directly into surface aquatic systems from the atmosphere (direct deposition) and Nr that is not either stored or removed as products on terrestrial systems eventually moves into aquatic systems (indirect deposition).

The area of an airshed generally greatly exceeds that of a watershed for a specific estuary or coastal region. For example, the airshed of the Baltic Sea includes much of western and central Europe (Asman, 1994; Hov et al., 1994), while the airsheds of the two largest U.S. estuarine ecosystems, the Chesapeake Bay and Albemarle-Pamlico Sound, are 15 to over 30 times the size of their watersheds (Dennis, 1997). Thus, the airshed of one region may impact the watershed and receiving waters of another, making eutrophication a regional-scale management issue (Galloway and Cowling, 2002; Paerl et al., 2002). Furthermore, atmospheric N inputs do not stop at coastal margins. Along the North American Atlantic continental shelf, atmospheric N inputs more than match riverine inputs (Jaworski et al., 1997; Paerl et al., 2002), underscoring the fact that N-driven marine eutrophication may require regional or even global solutions. Even in truly oceanic locations (e.g., Bermuda), North American continental atmospheric N emissions (reduced and oxidized N) are commonly detected and significant (Luke and Dickerson, 1987; Prospero et al., 1996). Likewise, islands in the North Pacific receive N deposition originating in Asia (Prospero et al., 1989).

Riverine and atmospheric "new" Nr inputs in the North Atlantic Ocean basin are at least equal and may exceed "new" Nr inputs from biological N_2 fixation (Howarth et al. 1996; Paerl and Whitall, 1999; Paerl et al. 2002). Duce et al. (2008) estimate that up to one-third of the oceans' external Nr supply enters through atmospheric deposition. This deposition leads to an estimated ~ 3% of new marine biological production and increased oceanic N_2O production. Schlesinger (2009) estimated that global atmospheric transport of Nr from land to sea accounts for the movement of almost one third of the annual terrestrial Nr formation. Therefore, our understanding of marine eutrophication dynamics, and their management, needs to consider a range of scales reflecting these inputs, including ecosystem, watershed, regional and global levels.

One example of shifting N inputs is the proliferation of intensive livestock operations in coastal watersheds, which has led to large increases and changes in chemical composition of nitrogenous compounds discharged to estuarine and coastal waters via runoff, groundwater, and atmospheric deposition (Paerl, 1997; Howarth, 1998; Galloway and Cowling, 2002). In general, coastal waters under the influence of these operations are experiencing increases in total N loading as well as a shift toward more reduced N (NH_4^+, organic N) relative to oxidized N (NO_3^-) (Galloway and Cowling, 2002; Howarth et al., 2002). These increases, combined with increases in hypoxia and anoxia in receiving waters, are leading to more NH_4^+-rich conditions, which will favor those algal groups best able to exploit this N form, including some harmful algal bloom (HAB) taxa (Paerl and Whitall, 1999; Paerl et al., 2007). Similarly, conversion of forest and agricultural lands to urban lands can alter landscapes and promote N loading to estuaries by increasing impervious pathways and removing natural landscape filters for Nr. Development also destroys wetlands, leading to more NO_3^--enriched conditions, potentially favoring plant taxa best able to exploit this N form.

A recent evaluation of decadal-scale changes of NO_3^- concentrations in ground water supplies indicates

Box 1: Hypoxia in the Gulf of Mexico

An example of a problem of excess Nr that moves from one part of the U.S. to another is the movement of Nr from the states that make up the Mississippi River drainage to the Gulf of Mexico (see discussion in Section 5.3.4). A hypoxic zone covers a significant area of the receiving bottom waters of the continental shelf of the northern Gulf of Mexico (details may be gleaned from U.S. EPA SAB, 2007). This is a seasonally severe problem that has persisted there for at least the past 20 years. Between 1993 and 1999 the hypoxia zone ranged in extent from 13,000 to 20,000 km^2 (Rabalais et al. 1996, 1999; Rabalais and Turner, 2001). The hypoxia is most widespread, persistent, and severe in June, July, and August, although its extent and timing can vary, in part because of the amplitude and timing of flow and subsequent nutrient loading from the Mississippi River Basin. The waters that discharge to the Gulf of Mexico originate in the watersheds of the Mississippi, Ohio, and Missouri Rivers (collectively described here as the Mississippi River Basin). With a total watershed of 3 million km^2, this basin encompasses about 40% of the territory of the lower 48 states and accounts for 90% of the freshwater inflow to the Gulf of Mexico (Rabalais et al. 1996; Mitsch et al., 2001; U.S. EPA SAB, 2007).

The December, 2007 report, *Hypoxia in the Northern Gulf of Mexico: An update by the EPA Science Advisory Board* (U.S. EPA SAB, 2007) determined that, "To reduce the size of the hypoxic zone and improve water quality in the Basin, the SAB Panel recommends a dual nutrient strategy targeting at least a 45% reduction in riverine total nitrogen flux (to approximately 870,000 metric tons/yr) and at least a 45% reduction in riverine total phosphorus flux (to approximately 75,000 metric tons/yr). Both of these reductions refer to changes measured against average flux over the 1980-1996 time period. For both nutrients, incremental annual reductions will be needed to achieve the 45% reduction goals over the long run. For nitrogen, the greatest emphasis should be placed on reducing spring flux, the time period most correlated with the size of the hypoxic zone."

that there has been a significant increase in nitrate concentrations in well water across the U.S. (Rupert, 2008). This study compared the nitrate content of 495 wells during 1988-1995 with nitrate content found during 2000-2004 as a part of the U.S. Geological Survey, National Water Quality Assessment Program. In a subset of wells, ground water recharge was correlated with historic fertilizer use and it was concluded that nitrate concentrations in ground water increased in response to the increase of N fertilizer use.

2.3.2. Storage of Nr within Terrestrial Environmental Systems

According to the nitrogen cascade conceptualization, terrestrial environmental systems are compartmentalized into agriculture, populated, and vegetated systems. Annual input of Nr is greatest in agricultural ecosystems (farmland, cropland, and grazed pastureland). Annual Nr inputs to agricultural ecosystems, using 2002 as the base year, include 9.8 Tg from synthetic fertilizer, 7.7 Tg from biological N fixation in crops (mainly soybeans), and 1.3 Tg from atmospheric deposition. Nr input into vegetated systems (mostly forested, but including non-cropland grasslands and other natural vegetation types as well) comes mostly from atmospheric deposition (3.2 Tg). Annual input of Nr into populated systems includes synthetic fertilizer application to urban turfgrass and recreational areas (~1.1 Tg), and atmospheric deposition 0.2 Tg.

Much of the annual Nr input into these terrestrial systems passes through, and is transferred within, terrestrial systems or atmosphere via NH_3, NO_x or N_2O, or aquatic environmental systems via NO_3^- and organic N leaching and runoff or NH_x and NO_y deposition.

The largest single reservoir of total N in the terrestrial environmental system is soil organic matter (SOM). Approximately 52,000 Tg C and 4,300 Tg N are contained in the upper 100 cm of soil in the 48 contiguous states (N is estimated from assumed C/N ratio of 12) (Lal et al., 1998). For comparison, the total above ground biomass of U.S. forests of these states contains ~ 15,300 Tg of C and ~ 59 Tg N (estimated using a C/N ratio of 261), and 15,500 Tg of soil organic matter carbon (SOM-C), and 1,290 Tg total N (estimated using a C/N ratio of 12) (U.S. EPA, 2007e). Most of this soil organic matter nitrogen (SOM-N) is bound within complex organic molecules that remain in the soil for tens to thousands of years. A small fraction of this SOM is mineralized, converted to carbon dioxide and Nr annually. The total N contained within above and below ground compartments isn't really of concern. What is of interest in addressing issues of Nr, is the change in N stored within the compartments of terrestrial systems. The pertinent question is whether N is being retained or released from long-term storage. The Committee evaluated estimates of annual change of N storage within important components of terrestrial systems. The Committee used Carbon stock information in Chapter 7 of the EPA Inventory of U.S. Greenhouse Gas Emissions and Sinks 1990-2005 (U.S. EPA, 2007e) to estimate N storage in U.S. terrestrial systems. Nitrogen stock change was determined by simply assigning a molar C/N ratio of 12 for soils and 261 for trees and making the appropriate conversions from C to N.

Agricultural systems

Croplands within the contiguous 48 states occupy ~ 368 million acres (149 million ha) (19%) of the 785 million ha of total land area. In 2002, 126 million ha of this cropland were cultivated (USDA NRCS, 2007). Croplands are generally found on well drained mineral soils (organic C content 1-6% in the top 30 cm). Small areas of drained organic soils are cultivated (organic C content of 10-20%) in mainly Florida, Michigan and Minnesota (EPA, 2007e). Organic soils lost ~0.69 Tg of Nr in 2002 while mineral soils accumulated ~1.5 Tg of Nr (Table 10). Much of the accumulation of SOM-C was due to the use of conservation tillage and high yielding crop varieties (U.S. EPA, 2007e). Losses of Nr from organic soils are due to mineralization of SOM and release of Nr input. In cultivated soils annual input of new Nr is approximately 9.7 Tg from fertilizer N, 1.1 Tg from livestock manure (recycled N), ~7.7 Tg from biological N fixation, and 1.2 Tg from atmospheric deposition. Assuming that loss of fertilizer N from the small area of organic soils is a minor fraction of the total, ~17% of N input from synthetic fertilizer, ~12% of total N input, is stored in cropland mineral soils annually.

According to the U.S. EPA National Greenhouse Inventory (U.S. EPA, 2007g) the net increase in soil C stocks over the period from 1990 through 2005 was largely due to an increase in annual cropland enrolled in the Conservation Reserve Program, intensification of crop production by limiting the use of bare-summer fallow in semi-arid regions, increased hay production, and adoption of conservation tillage (i.e., reduced- and no-till practices). It is clear that conversion of marginal crop land to CRP stores C and reduces erosion and nitrate leaching. Likewise, use of soil conservation tillage, as opposed to conventional tillage with a plow or disk, reduces erosion. However, the impact of conservation tillage on soil C storage and Nr losses due to leaching or denitrification are much less certain. For example, although the EPA estimates shown in Table 10 assume that no-till crop production results in net carbon sequestration, recent publications indicate that no-till cropping practices do not result in net carbon sequestration (Verma et al., 2005; Baker et al. 2007; Blanco-Canqui and Lal, 2008). Therefore, the estimates of soil C and N storage in mineral soils in Table 10 that were derived from U.S.EPA, (2007b) need to be reconsidered. These new studies and that of David et al. (2009) suggest that organic C conservation by reduced tillage practices has been overestimated because soil sampling and analysis has been confined to the top 30 cm of soil when the top meter of soil needs to be considered. Baker et al. (2007) and Verma et al. (2005) also show that long-term, continuous gas exchange measurements have not detected C gain due to no-till practices. They concluded that although there are other good reasons to use no-till, evidence that it promotes C sequestration is not compelling. These findings highlight the need for appropriate assessment of ecosystem N storage in order to confirm or disprove this Committee's conclusion that only a small part of annual Nr input is stored in agricultural lands, forests, and grasslands.

Populated systems – urban lands

Populated or "developed land" (developed land is the terminology used by the U.S Department of Agriculture's Natural Resources Conservation Service [NRCS]) occupied ~106 million acres (42.9 million ha) of the U.S. land area in 2002. This equates to approximately 5.5% of the U.S. land area (USDA NRCS, 2007). The EPA Inventory of U.S. Greenhouse Gas Emissions and Sinks (U.S. Environmental Protection Agency, 2007e) indicates that urban areas cover over 4.4% of the land area with tree canopy covering 27.1% of the urban area. The tree-covered area constitutes approximately 3% of total tree cover in the continental U.S. If the NRCS value of 42.9 million ha is used, then trees cover ~27.9 million acres (11.3 million ha) of urban land in the contiguous 48 states. Another ~ 35.1 million acres (14.2 million ha) of land is covered by turf grass in parks, golf courses, and lawns. In both urban forests and turf grass, Nr storage is dependent upon the age of the trees or turf. In young (pre-steady state) systems N is being accumulated, while at steady state no net change occurs. Some areas may be degrading and actually losing biomass and returning N to the environment. EPA does not estimate carbon changes in turf grass, but does estimate changes in carbon storage in urban forests (U.S. EPA, 2007e). Urban trees sequestered an estimated net 22 Tg of carbon and 0.12 Tg of N in 2002 (assuming that urban trees are mainly hardwood having a C/N ratio of 186) (U.S. EPA, 2007e). Annual fertilizer N input into the urban landscape is approximately 10% of total fertilizer N consumption in the U.S. (U.S. EPA, 2007e), or ~1 Tg of N in 2002. Another 0.2-1.0 Tg N is deposited from the atmosphere. In some areas deposition can be disproportionately high due to locally high NO_y concentrations. Storage of ~0.12 Tg N in urban forests constituted approximately 3% of Nr input annually.

Vegetated systems – forests and grasslands

Forests cover approximately 164 million ha, ~21% of the land area of the contiguous 48 states (USDA NRCS, 2007). The forest carbon stocks analysis by EPA (U.S EPA, 2007e) is based on state surveys that are conducted every 1 to 10 years. Annual averages are applied to years between surveys. To determine changes in C related to the rate of tree growth, Birdsey (1992) estimated that there were 52,500 Tg of C above and below ground in U.S. forests; soil contained 59% of total C, 9% was in litter, and 5% in tree roots. The EPA estimate for 2002 was 43,600 Tg of C. To estimate N storage based on EPA data (U.S. EPA, 2007e), the Committee has assumed that forests are 85% softwood and 15% hardwood with an average C/N ratio of 261. These estimates indicate that forests and forest products stored ~0.43 Tg of N in 2002 (Table 10).

Grasslands, including rangelands and pasturelands, occupy approximately 213 million ha (27.1%) of the contiguous 48-state land area. The NRCS divides these grasslands into pastureland (48.2 million ha) and rangeland (164 million ha). Pastureland is managed (it may be fertilized and mown) and rangeland is managed only to the extent that livestock grazing intensity is regulated on the land used for such grazing. Changes in the N status of grasslands are dependent upon changes in soil organic matter as the above ground biomass produced annually is either consumed by livestock or decomposed in the field. Soil organic C stocks were estimated using the Century biogeochemical model and data used were based upon the NRCS/National Resources Inventory (NRI) survey (U.S. EPA, 2007e). Changes in soil N content were estimated using a C/N ratio of 12. Nitrogen input into rangelands is generally only from atmospheric deposition, which contributes 1.9 Tg N each year to range production. Rangeland tends to be in relatively remote areas where atmospheric Nr deposition is low.

Collectively, forests and grasslands stored ~0.74 Tg of N in 2002. Much of the soil N storage in grasslands is a result of conversion of croplands to grasslands, mainly due to the conservation reserve program. Forest soils

Table 10: Net annual change in continental U.S. croplands soil N and C, forest C and N, and grasslands C and N in 2002

	C	N
Cropland		
Cropland remaining cropland		
Mineral soil	17	1.4*
Organic soil	-8.3	-0.69
Land converted to cropland	0.8	0.067
Total	**9.5**	**0.80**
Forests		
Forests and harvested wood products		
Above ground biomass	85	0.32
Below ground biomass	16	0.063
Dead wood	9.1	0.035
Litter	7.2	0.028
Soil organic matter	-2.8	-0.23
Harvested Wood	59	0.22
Total	**174**	**0.44**
Grasslands		
Grasslands remaining grasslands		
Mineral soil	-0.8	-0.067
Organic soil	-1.3	-0.11
Lands Converted to Grasslands	5.8	0.48
Total	**3.7**	**0.31**
US Total C & N Storage in 2002	**187**	**1.5**

Measurements in Tg. Negative sign indicates a decrease in storage; positive number indicates increase in storage. Soil C/N ratio = 12; wood C/N = 261. C storage numbers were obtained from U.S. EPA, 2007e.* See previous discussion of soil organic matter accumulation in croplands.

appear to be losing N while overall N storage in forests is from accumulation in above-ground biomass and that that remains in forest products that are stored for long periods.

Summary of estimates of Nr stored in terrestrial systems in 2002

Although our estimate of N storage in terrestrial systems is highly uncertain, annual N storage is likely relatively small. Approximately 1.5 Tg N was stored in terrestrial systems of the contiguous 48 states in 2001 (Table 10). Soils were the largest reservoir with croplands (0.8) and grasslands (0.3) sequestering most of the N. Annual storage in agricultural, grassland and forest soil and in forest biomass is approximately 6 to 10% of annual Nr input. Estimated total Nr input from synthetic fertilizer, biological N fixation, and atmospheric deposition into terrestrial systems within the contiguous 48 states in 2002 was ~32 Tg. All of the input and outflow numbers are highly uncertain, but N loss through denitrification appears to be the major loss mechanism. Storage in soils and trees appears to account for only a small portion of the annual N input while apparent loss through denitrification dominates the budget. This is discussed in Section 2.3.3 of this report and in a recent global Nr review by Schlesinger (2009). Some small fraction is re-volatilized and exported from the continent.

2.3.3. Areas of Uncertainty in Nr Transfer and Transformation

In considering Nr transfers and transformations in and between the environmental systems of the nitrogen cascade, the Committee has encountered a number of areas where quantities or flows of Nr are highly uncertain. All of these areas need attention from EPA in conjunction with other federal and state agencies and universities. Although most of the following points have been highlighted in various findings and recommendations within other chapters of this report, we feel the need to highlight the following areas:

- Total denitrification in animal feeding operations, in soils, and in aquatic systems needs to be quantified along with all gaseous products produced and released to the atmosphere during nitrification/denitrification. These gases include NOx, N2O and N2.
- The amount of Nr transferred to each environmental system as dry deposition needs to be quantified and monitored.
- The fraction of NOy in the form of organo-nitrates and other organic nitrogen species is poorly quantified, but may play a major role in air quality and Nr cycling.
- Rates and amounts of ammonia emissions from fertilized soils and animal feeding operations need to be quantified and the fate of this ammonia determined.
- The annual change in N storage in soils (agricultural, forest, grassland and urban areas) needs to be quantified in conjunction with the change in carbon.

These areas of high uncertainty are highlighted because very little information exists in some of the areas. In other areas, such as denitrification and the relative release of N_2O from soils and aquatic systems, the sparse data are highly variable and this makes developing meaningful guidelines for control difficult. An analysis of Nr input and fate in 16 watersheds in the U.S. is provided as an example to illustrate how the inputs and fate of Nr can be evaluated for a large watershed.

Input and fate of Nr in 16 watersheds in the northeast United States

There are no comprehensive data available to assess the transfer and transformations in and between the atmosphere, terrestrial systems (agriculture, populated, and vegetated systems) and aquatic systems nationally. Determining a national N budget is a priority research area. As there are no national data available, an example analysis of Nr input and fate in 16 watersheds in the northeast U.S. (for which data are available) is used to show an evaluation of the inputs and fate of Nr for a large watershed (Van Breemen et al., 2002).

The watersheds in this study encompass a range of climatic variability, Maine to Virginia. The watersheds are a major drainage to the coast of the North Atlantic Ocean. Using data from the early 1990s, Boyer et al. (2002) reported the quantification of N inputs to each watershed from atmospheric deposition, N fertilizers, biological N fixation, and import of N in agricultural products (food and feed). They compared inputs with N losses from the system in riverine export. As a part of the same study, Van Breemen et al. (2002) analyzed the fate of N inputs to these watersheds and developed budgets for each watershed. The total area of the watersheds was 32,666 square km with land use categories of forest (72%), agricultural (19%), urban (3%), wetlands (5%), and 1% other uses. The Nr input into the watersheds (using weighted averages for all 16 watersheds) was 3,420 kg per square km per year. Figure 14 shows the Nr sources and the estimated fate of this Nr as a per cent of the weighted average Nr input.

Van Breemen et al. (2002) indicate that Nr inputs and storages and losses were well correlated ($R^2 = 0.98$). Denitrification in landscape soils is the most uncertain estimate because rates are calculated by difference between total inputs and outputs so they accumulate errors from all estimates. The denitrification loss term may also reflect the change in N storage in groundwater. The net storage of N in the soil (18% of total storage and losses) indicates that there is a non-steady state condition in the soil. Increasing storage of Nr on land implies that drainage and denitrification exports of Nr are likely to increase when a new steady-state condition is reached.

These data suggest that Nr research needs to focus on understanding the “denitrification” loss term in this analysis. The losses occur in the terrestrial landscape, before Nr

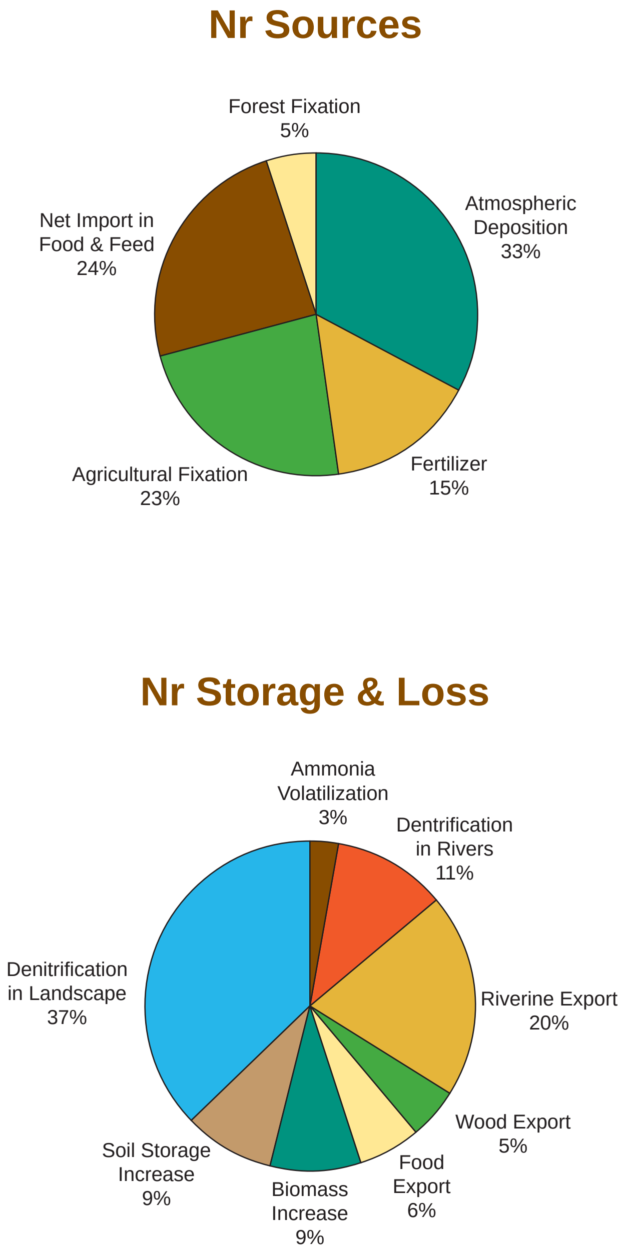

Figure 14: Nr input and loss from 16 watersheds in the northeast United States

Source: Van Breeman et al., 2002 (Figure 7, p. 289). Reprinted with permission; copyright 2002, Springer Science+Business Media B.V.

enters the river. Where do these losses occur, within the agricultural field, in drains and ditches near the agricultural field, in riparian areas, or wetlands? Understanding this term may help in the management of Nr in watersheds to decrease nitrate movement into aquatic systems as well as to limit N_2O emissions to the atmosphere.

The Van Breemen et al. (2002) study also estimated that approximately 30% of N input was exported to the rivers and about two thirds (20% of total N input) of this N was exported to coastal waters by rivers. The remaining one-third (11% of total N input) was considered to have been denitrified in the rivers. These examples also demonstrate that Nr in the atmosphere, terrestrial systems and aquatic systems are not separate and must be considered collectively. Atmospheric deposition is a variable but important input into aquatic systems that contributes to Nr enrichment problems. Aquatic and terrestrial systems process this Nr and return other Nr gases (NH_3, NO_x and N_2O to the atmosphere). Nr from terrestrial systems impacts both the atmosphere and aquatic systems through emission of NH_3, NO_x, N_2O and leaching and runoff of NO_3^-.

Finding 10: Denitrification of Nr in terrestrial and aquatic systems is one of the most uncertain parts of the nitrogen cycle. Denitrification is generally considered to be a dominant N loss pathway in both terrestrial and aquatic systems, but it is poorly quantified.

Recommendation 10: *EPA, USDA, DOE, and universities should work together to ensure that denitrification in soils and aquatic systems is properly quantified, by funding appropriate research.*

3

Impacts of Nr on Aquatic, Atmospheric, and Terrestrial Ecosystems

This chapter summarizes knowledge of the impacts of Nr on freshwater, coastal, atmospheric, and terrestrial ecosystems. Table ES-1 in the Executive Summary provides a more detailed presentation of the quantitative extent of Nr impacts on the environment.

3.1. Impacts on Drinking Water, Human Health, and Freshwater Biota

A detailed presentation on the impacts of Nr on various aquatic systems is presented in Appendix G. The EPA's Office of Water (U.S. EPA, 2007b) has noted the following impacts caused by excessive Nr in aquatic systems:

- Excessive nutrients (N and P) can cause negative ecological impacts to water bodies on a national scale by stimulating harmful algal blooms.
  - Algal blooms block sunlight and result in the destruction of submerged aquatic vegetation which serves as critically important habitat and food for many organisms.
  - Algal blooms eventually die off and consume dissolved oxygen from the water column which can lead to die off of aquatic organisms.
  - One result of algal blooms is decreased biological diversity and populations, including smaller populations of game and commercial fish.
  - Some blooms, considered harmful algal blooms or HABs, have a toxic effect on living organisms and are disruptive of ecosystem structure and transfer of energy to higher trophic levels.
- Excessive nutrients also pose public health risks.
  - Algal blooms can cause taste and odor problems in drinking water.
  - Hazardous algal blooms can cause respiratory distress and neurological problems in swimmers.
  - Excessive nitrates can cause "blue baby syndrome."
- Nutrient pollution is occurring at a national scale and has not been completely addressed.
  - 49 states and 4 territories have 303(d) listings due to nutrients, and about 50% of the states have greater than 100 water quality impairments due to nutrients.
  - Over 10,000 impairments are a result of nutrient pollution.

Finding 11: There is growing recognition of eutrophication as a serious problem in aquatic systems (NRC, 2000). The last comprehensive National Coastal Condition Report was published in 2004 (U.S. EPA, 2004) and included an overall rating of "fair" for estuaries, including the Great Lakes, based on evaluation of more than 2,000 sites. The water quality index, which incorporates nutrient effects primarily as chlorophyll-a and dissolved oxygen impacts, was also rated "fair" nationally. Forty percent of the sites were rated "good" for overall water quality, while 11% were "poor" and 49% "fair."

Recommendation 11. *The Committee recommends that EPA develop a uniform assessment and management framework that considers the effects of Nr loading over a range of scales reflecting ecosystem, watershed, and regional levels. The framework should include all inputs related to atmospheric and riverine delivery of Nr to estuaries, their comprehensive effects on marine eutrophication dynamics and their potential for management.*

Finding 12: Meeting Nr management goals for estuaries, when a balance should be struck between economic, societal, and environmental needs, seems unlikely under current federal law. Enforceable authorities over nonpoint source, stormwater, air (in terms of critical loads), and land use are not adequate to support necessary Nr controls. Funding programs are presently inadequate to meet existing pollution control needs. Furthermore, new technologies and management approaches are required to meet ambitious Nr control needs aimed at restoring national water quality.

Recommendation 12. *The Committee recommends that EPA reevaluate water quality management approaches, tools, and authorities to ensure Nr management goals are attainable, enforceable, and the most cost-effective available. Monitoring and research programs should be adapted as necessary to ensure they are responsive to problem definition and resolution, particularly in the development and enhancement of nitrogen removal technologies and best management practices, and continue to build our level of understanding and increase our ability to meet management goals.*

3.2. Impacts of Airborne or Atmospherically Deposited Nr on Human Health and Ecosystems

Six major atmospheric effects are associated with increased NO_x and NH_3 emissions, and two with N_2O emissions (Galloway et al., 2003). For NO_x and NH_3 emissions, the effects are:

- Decreases in atmospheric visibility caused by fine PM
- Elevated ozone concentrations that enhance the greenhouse potential of the atmosphere
- Serious ozone and fine particulate matter impacts on human health (Pope et al., 1995; Pope, 2000a,b ; Brook et al., 2003; Brunekreef et al., 2005; Pope, 2009)
- The important role that NH_3 plays in the direct and indirect effects of aerosols on radiative forcing and thus on global climate change (Penner et al., 1991; Seinfeld and Pandis, 1998; Lelieveld et al., 2001; Myhre, 2009)
- Decreased productivity of crops, forests, and natural ecosystems caused by ozone deposition
- Atmospheric deposition of NH, NH_3, NO_y, and organic forms of Nr that can contribute to ecosystem acidification, fertilization, and eutrophication

For N_2O, the effects are the greenhouse effect in the troposphere and O_3 depletion in the stratosphere.

3.3. Impacts of Nr on Terrestrial Ecosystems

As previously discussed, in many terrestrial ecosystems the supply of biologically available Nr is a key factor controlling the nature and diversity of plant life, and vital ecological processes such as plant productivity and the cycling of carbon and soil minerals. Human activities have not only increased the supply but enhanced the global movement of various forms of nitrogen through air and water. Appendix H presents information on the impact of Nr saturation on ecosystem function.

The primary source of excess Nr for most unmanaged terrestrial ecosystems is atmospheric deposition. This additional Nr causes a wide variety of sometimes beneficial effects (increased growth and productivity of forests, natural grasslands, and crops planted in nutrient deficient soils) and also sometimes adverse effects on terrestrial and aquatic ecosystems in many parts of the U.S. Forests and grasslands exposed to excess Nr can respond in numerous ways. General effects include the following (Woodman and Cowling, 1987; Cowling, 1989; Garner et al., 1989; Cowling et al., 1990; Vitousek et al., 1997a,b; Cowling et al., 2002):

- Increased productivity of forests soils, most of which are Nr-limited throughout the U.S. Nr deficiency of forest soils has been most fully quantified for pine forests in 14 southeastern states.
- Acidification of forest soils leading to decreased availability of nutrient cations including calcium, magnesium, and potassium and aluminum toxicity, established most clearly in the eastern U.S. and both central and northern Europe.
- Nr saturation of forest soils (which results in increased Nr release to the water draining the soils), presently occurring mainly in high-elevation forests of the eastern U.S. and southeastern Canada.
- Ozone-induced predisposition of forest trees to damage by fungal diseases and insect pests, most clearly established in the case of root disease and bark beetles in the pine forests of southern California.
- Ozone-induced inhibition of photosynthesis in both softwood and hardwood tree species most clearly established in controlled exposure studies in both the U.S. and Europe at ambient concentrations of ozone above 60 ppb. Such concentrations occur frequently throughout the eastern U.S. and southeastern Canada.
- Ozone-induced direct injury to foliage, most clearly established in the case of “emergence tip burn” in eastern white pine.
- Acidification-induced decrease in frost hardiness of high-elevation conifer forests, most clearly established in the case of red spruce in the northeastern U.S.
- Acidification-induced alteration of beneficial symbiotic relationships in forest soils, especially mycorrhizae, most clearly established in both northern and central Europe.
- Biodiversity losses in natural grasslands and forest areas caused by Nr-induced decreases in abundance of Nr-limited tree and grass species and replacement by Nr-loving weed species, most clearly established in both Minnesota and California, and even more vividly in The Netherlands.
- Decreases in visibility and increased haziness of the atmosphere at scenic vistas in national and state parks and wilderness areas.
- More leaching of Nr to aquatic systems via both groundwater and surface runoff—a cascade effect.

4

Metrics and Implications for Risk Reduction Strategies for Reactive Nitrogen

It is important to develop risk reduction strategies for reactive nitrogen that take into consideration the ways in which Nr is introduced and transformed in the environment. This chapter reviews current and historical measurement and risk reduction activities for Nr and provides specific Committee findings and recommendations.

4.1. Measurement of Nr in the Environment

Although nitrogen is among the most abundant elements on earth, only a small fraction, Nr, is responsible for impacts on the environment. Most regulations focus narrowly on specific chemical forms of nitrogen as they affect media- or site-specific problems, setting limits or specifying control technologies without regard to the ways in which N is transformed once introduced into the environment. Measurement methods are typically expressed in terms of mass loadings or concentrations of a particular form of N (e.g., ppm NO_x, mg/L total NH_x, or kg/ha of NO_3^-).

Finding 13: The Committee finds that there is a need to measure, compute, and report the total amount of Nr present in impacted systems in appropriate units. What is measured influences what we are able to perceive and respond to; in the case of Nr, it is especially critical to measure total amounts and different chemical forms, at regular intervals over time.

Recommendation 13: *The Committee recommends that EPA routinely and consistently account for the presence of Nr in the environment in forms appropriate to the medium in which they occur (air, land, and water) and that accounting documents be produced and published periodically (for example, in a fashion similar to National Atmospheric Deposition Program [NADP] summary reports). The Committee understands that such an undertaking will require substantial resources, and encourages the Agency to develop and strengthen partnerships with appropriate federal and state agencies and private-sector organizations having parallel interests in advancing the necessary underlying science of Nr creation, transport and transformation, impacts, and management.*

4.2. Consideration of Nr Impacts in Risk Reduction Strategies

Historical measurement and impact categories

The types of impacts of Nr in the environment are dependent on three general factors: the sources of Nr, the types of media impacted, and Nr chemical forms. The magnitude of effects depends on loading and the nature of the system impacted. As illustrated in Figure 1, the impacts of a given source of Nr can be multiple as N is transformed in the environment and transported among ecosystem components. The nitrogen cascade provides a comprehensive framework for understanding the role of Nr in the earth's ecosystems and establishes a framework for developing and implementing management methods through which beneficial effects can be enhanced while minimizing detrimental impacts.

A management paradigm in which various approaches are used to limit environmental impacts to "acceptable" levels of risk is a useful concept for understanding the environmental impacts that Nr can have. For this purpose, impacts are divided into several general categories within which various contaminants have a direct correlation with damage. Once the nature and type of impacts are recognized, the risks should be characterized quantitatively, if possible. This information would then be used, along with other considerations such as economic, social and legal factors, to reach decisions regarding risk reduction strategies and the need for and practicability of implementing various risk reduction activities. The regulation of Nr in the environment by EPA follows an impact-by-impact approach which, with few exceptions, examines specific N forms in either aquatic, atmospheric, or terrestrial systems. As previously discussed, the principal regulatory authority pertaining to nitrogen is derived from the CWA and the CAA, although other legislation such as the Energy Independence and Security Act (EISA), and the Endangered Species Act (ESA) contain provisions that could result in regulatory actions that affect nitrogen management.

Historically, EPA environmental protection programs have addressed impacts of Nr such as climate change, eutrophication, ecotoxicity, human health (cancer and non-cancer), acidification, smog formation, and stratospheric ozone depletion, among others (Bare et al., 2003). Within these categories it is sometimes possible to express end points in terms of collective metrics, such as is done with greenhouse gases in the form of carbon dioxide equivalents, or acidification as H^+ equivalents. This approach has the considerable advantage of defining a straightforward framework within which environmental standards can be derived that are protective of human health and the environment – EPA's principal mission. This approach also encourages evaluation of damage from multiple sources, as long as the characterization metric used is genuinely representative of the impact of a given contaminant. Thus, for example, the total impact of acidic gases such as SO_2 and NO_x on the acidification of watersheds can be expressed as a common metric. However, metrics for human health are generally not as simple to characterize nor are the appropriate end points; thus, the mechanism of toxicity, number of individuals affected, value of lost workdays, medical treatment costs, and value of human lives lost may all be used.

Ecosystem functions and services

A complementary approach to classical impact characterizations is the use of ecosystem "service" and "function" categories, in which the impairment of a specific service provided by one or more ecosystems or impairment of an ecological function by causative contaminant emissions is assessed (Costanza, 1997; Millennium Ecosystem Assessment, 2003). Such an approach is inherently attractive because of its basis in scientific reality, i.e., the health of humans is inextricably linked to the health of the environment. Less clear, in some cases, are ways in which to measure and monitor such impacts and account for the effects of a complex array of factors and stressors that contribute to or damage ecosystem service, function, and health. Table 11 provides examples of ecosystem services and corresponding functions.

The use of ecosystem services in a regulatory context would be a different approach for the EPA, one with considerable potential, but one for which experience is currently lacking. In comparison to the available data on reactive nitrogen usage, little is known about the response of ecosystems and ecosystem services to reactive nitrogen loads. This is discussed more fully in Sections 4.5, 4.6 and Appendix D on critical loads. In this context the Committee

Table 11: Ecosystem service and corresponding function categories

Ecosystem Service	Ecosystem Function
Gas regulation	Regulation of atmospheric chemical composition
Climate regulation	Regulation of global temperature, precipitation, and other biologically mediated climatic processes at global, regional, and local levels
Disturbance regulation	Capacitance, damping, and integrity of ecosystem response to environmental fluctuations
Water regulation	Regulation of hydrologic flows
Water supply	Storage and retention of water
Erosion control and sediment retention	Retention of soil within an ecosystem
Soil formation	Soil formation processes
Nutrient cycling	Storage, internal cycling, processing, and acquisition of nutrients
Waste treatment	Recovery of mobile nutrients, and removal or breakdown of toxic compounds
Pollination	Movement of floral gametes
Biological control	Trophic dynamic regulation of populations
Refugia	Habitat for resident and transient populations
Food production	That portion of gross primary production extractable as food
Raw materials	That portion of gross primary production extractable as raw materials
Genetic resources	Sources of unique biological materials and products
Recreation	Providing opportunities for recreational activities
Cultural	Providing opportunities for noncommercial uses

Source: Costanza et al., 1997 (Table 1, p. 254). Reprinted with permission; copyright 1997, Nature Publishing Group.

supports plans by the EPA to incorporate research on the services concept, focusing on Nr as the suite of contaminants of interest, into its future ecological research plan (U.S. EPA, 2009a). EPA's Ecological Research Plan was reviewed by the Science Advisory Board (U.S. EPA SAB, 2008). More recently, the Science Advisory Board completed a self-initiated study on "Valuing the Protection of Ecological Systems and Services" (U.S. EPA SAB, 2009). This report explores the concept of ecosystem services as a basis for regulatory action and presents a roadmap for implementing this approach.

Economic measures and impacts

It is also possible to translate the effects of Nr into economic terms. Two economic measures that are often used are the dollar costs of damages and the cost of remediation or substitution. Another important economic metric is the cost/ton of remediation for each form of Nr. Damage costs do not always scale as tons of Nr released into the environment. If damage costs rather than tons of nitrogen were utilized as a metric, the full implications of the cascade and the setting of priorities for intervention might differ.

It is important to note that the choice of metric used in assessing impacts may play an influential role in what and how one manages. Air and water protection laws state that the goal is "to protect human health and the environment." Yet, there is no generally agreed-upon common metric for measuring the full range of effects (which are complex and often unknown) or for setting priorities in the establishment or implementation of policies.

As noted above, there are multiple metrics for measuring Nr or any other agent in the environment. The most common metric utilizes quantitative measures of the total amount of Nr (and any of its specific chemical forms) in different environmental reservoirs and the mass flux between them. But while providing common units, typically mass or concentration, these measures do not distinguish the relative societal costs of health or environmental consequences of reactive nitrogen of different forms or places in the cascade. While not all damages can be turned into economic costs, and the costs of some damages have not been quantified, enough of the major damages can be quantified economically to provide a useful complementary metric for decision-making. (See the Chesapeake Bay example in Box 2.)

The advantage of monetizing damages is that it reflects an integrated value that human society places on lost ecosystem goods and services in common currency and illustrates the cascading costs of damages as Nr changes form and moves between different parts of the ecosystem. In addition, human health implications can also be included as the cost of health care treatment, lost work days and other aspects of morbidity and mortality (e.g., economic value of lives lost). A third metric is to look at morbidity and/or mortality separately and not monetize them with a cost value. Of course a concern, particularly with respect to the economic metric, is that there are a number of ecosystem services that arguably cannot be easily monetized, for example the loss of biodiversity and those ecosystem functions that are affected by climate change or other stressors. Ecosystem services considered to be regulating and supporting are particularly difficult to fit into an economic metric. It is thus essential that a variety of complementary metrics be used to assess the impact of anthropogenic Nr on the environment and human well being.

There is value in each of the ways that N metrics are expressed. Traditional categories provide a readily adaptable framework for regulation, while ecosystem service and function-based categories provide a richer context for stating the complex connections among Nr inputs and transformations and their impacts on ecosystem health and human well-being. Dollar-based metrics provide a means of identifying those effects that have the greatest impacts and costs to society.

Finding 14: The Committee finds that reliance on only one approach for categorizing the measurement of Nr is unlikely to result in the desired outcome of translating N-induced degradation into the level of understanding needed to develop support for implementing effective Nr management strategies.

Recommendation 14: *It is, therefore, recommended that the EPA consider the impact of different metrics and examine the full range of traditional and ecosystem response categories, including economic and ecosystem services, as a basis for expressing Nr impacts in the environment, and for building better understanding and support for integrated management efforts.*

4.3. Water Quality Regulation and Management

Aquatic thresholds

In aquatic ecosystems, thresholds at which excess Nr becomes a problem can be expressed as a management goal such as a total maximum daily load (TMDL) or as a critical load (CL). Under the authority of the CWA, EPA has developed guidance for establishing numeric nutrient criteria on an eco-regional basis for lakes and reservoirs, streams and rivers, estuaries and coastal waters, and wetlands. EPA has proposed specific numbers for lakes and reservoirs and rivers and streams and protocols for developing criteria for estuaries and wetlands. Each state is advised to go through an assessment to determine the best methodology for implementing numeric criteria (U.S EPA, 2000c, 2000c, 2001b, 2007e). These criteria will identify impaired waterbodies for which TMDLs may be required.

The second type of threshold available for aquatic ecosystems is the critical load (CL). Unlike the TMDL, the CL (in the U.S.) has no regulatory framework but rather sets the threshold of Nr loading at which negative

Box 2: Economic Impact and Metrics for Chesapeake Bay and Its Watershed

Recently, the N cycle and the implications of the reactive nitrogen cascade were translated into economic terms for the case of Chesapeake Bay (Moomaw and Birch, 2005). This approach has recently been updated with more recent data, and the economic and health impacts of different forms of Nr in multiple ecosystems and media have been estimated using better modeling methods (Birch et al., 2011). As an illustration, each of these metrics is shown as a percentage of Nr fluxes in the Chesapeake Bay water and air shed in Figure 15. Abatement costs are summarized in Table 12. Atmospheric emissions account for 37% of Nr entering the watershed, but they account for 75% of the dollar damages and 76% of the mortality (U.S. EPA, 2005c). Mitigation costs per tonne of atmospherically released Nr are the lowest among the three sources. Additions of Nr to terrestrial ecosystems add 60% to the system, but contribute only 24% of the damage costs and 24% of the mortality, and have the highest mitigation costs. Freshwater releases, the second most expensive to mitigate, account for the smallest portion of Nr contributions to the system by any of the metrics considered: only 4% of the Nr, 2% of the cost damages, and none of the mortality losses (Birch et al., 2011). Costs of Nr damage and health metrics provide additional economic measures of the cost effectiveness of actions to reduce a metric ton of Nr.

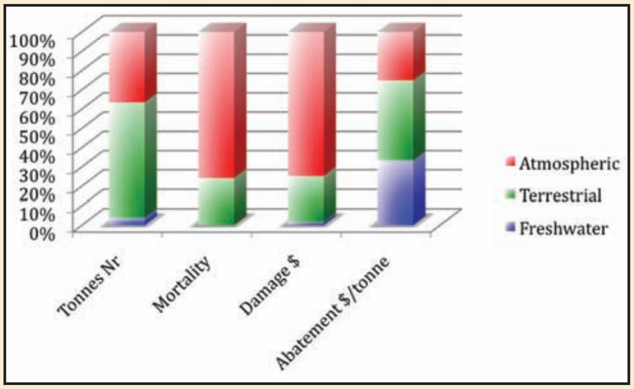

Figure 15: Relative importance of all reactive nitrogen sources released into atmospheric, terrestrial, and freshwater media within the Chesapeake Bay Watershed utilizing four different metrics

Source: Birch et al., 2011 (Figure 3, p. 173). Reprinted with permission; copyright 2011, American Chemical Society.

The metrics of damage cost and mortality (morbidity shows a similar pattern to mortality, but is only one-tenth the damage cost) indicate that controlling emissions of NO_x from combustion and industrial processes produces greater gains in protecting human health and the environment than does reducing other Nr releases, though the two sources are comparable in terms of reactive forms of Nr released to the watershed. This difference occurs because emissions to the air cascade through more parts of the watershed ecosystem than releases directly to the Bay. If human health effects are monetized, then the economic gains are even greater from reducing atmospheric emissions (see Figure 16).

Figure 16 is a scatter plot of all quantifiable damage costs (including health impacts) relative to metric tons of Nr showing the significant difference in emphasis of the two metrics. Note that direct additions to the environment from agriculture are about 370,000 tonnes Nr/year, and cause $1.7 billion worth of damage. Emissions of NO_X from mobile sources represent only 180,000 metric tons Nr/year but cause nearly $4.4 billion in damages each year, of which $108 million is attributable to nitrate loading of the Chesapeake Bay, $3.9 billion to human morbidity and mortality, and the remainder to other forms of damage, such as crop and commercial forest damage. Hence the releases of Nr into the airshed from mobile sources, which are only half the amount of agricultural releases to the watershed, cause more than 2.5 times the economic damage of environmental additions from agriculture. This integrated inclusion of atmospheric, terrestrial, and aquatic additions of Nr is not reflected in today's regulations

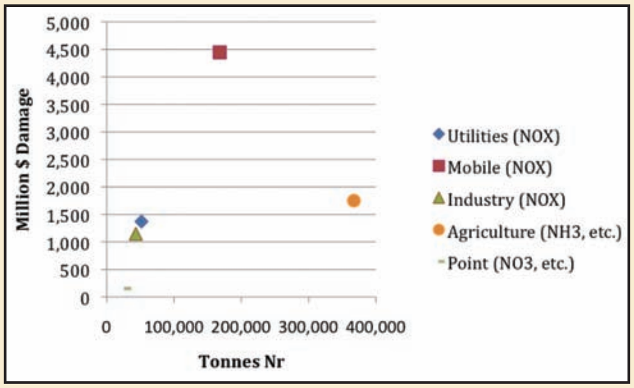

Figure 16: Quantified damage costs (including health impacts) relative to metric tons of reactive nitrogen in Chesapeake Bay Watershed

Source: Birch et al., 2011 (Figure 4, p. 173). Reprinted with permission; copyright 2011, American Chemical Society.

Marginal abatement costs per metric ton Nr by source into each of the three media are provided in Table 12, and demonstrate that the least costly abatement cost per metric ton of Nr also comes from atmospheric emission controls. While most legislation constrains how cost for remediation can be considered, it is useful to know where the lowest cost options lie in setting priorities.

These multiple metrics provide several ways of looking at the nitrogen cascade and its impact on human health and the environment. However, there are many impacts that remain unaccounted for in any of these metrics. Some impacts might be quantified, but the necessary data have yet to be collected. Economic losses due to damage to commercial fisheries in the Bay are an example that is likely to be significant but has not yet been quantified. Similarly, economic losses due to climate change and ozone depletion from N_2O emissions have not been fully evaluated. Impacts such as loss of biodiversity cannot be readily quantified at all, so it is desirable to consider a set of qualitative and non-quantified metrics in addition to the quantitative ones.

Other parts of the country such as the Mississippi valley or the Central Valley of California are expected to show very different patterns of cost damages, with terrestrial and freshwater emissions causing proportionally higher damage costs, and emissions to the atmosphere causing a lower percentage of damages. But those very differences would assist EPA and the generators of those emissions in setting priorities for mitigation.

It is important to recognize that Nr is not the only stressor that can affect both human and environmental health. Researchers are challenged to comprehensively understand cause-and-effect relationships in a complex environment and to balance management actions and costs to ensure that risk-minimizing management strategies are effectively implemented.

As these multiple metrics indicate, decisions about which fluxes of Nr to mitigate depend upon which metric is utilized. The cascading economic costs of damage highlight the importance of regulating air emissions because of their impacts on human heath as well as their large contribution to the degradation of Chesapeake Bay water quality. Hence, if one is interested in reducing water impacts of Nr, the total reduction of damage may rely nearly as much on stricter enforcement of the Clean Air Act as the Clean Water Act. This challenges our traditional approach to regulation, but that is a consequence of comprehensively examining Nr guided by the nitrogen cascade.

Table 12: Marginal abatement cost per tonne of Nr by source

Location in the N cascade where emitted	Source/pollutant	Abatement cost per tonne of Nr
Air	Electric utilities/NO_x [14]	$4,800
	Industrial/NO_x [15]	$22,000
	Mobile sources/ NO_x [16]	$14,000
	Non-agricultural/NH_3	No estimate
Land	Agriculture/nitrate [17]	$10,000
	Urban and mixed open land uses/nitrate [17]	$96,000
Fresh water	Point sources/nitrates [17]	$18,000

impacts have been documented. Based extensively on European work, CLs for aquatic ecosystems are Nr inputs on the order of 2-15 kg N/ha/yr (Bobbink et al., 2010). There are numerous locations within the U.S. where deposition to surface waters falls within this range.

Water quality standards

Section 303 of the CWA requires states to adopt water quality standards and criteria that meet the state-identified designated uses (e.g., uses related to "fishable" and "swimmable") for each waterbody. Specifically, "a water quality standard defines the water quality goals of a water body, or portion thereof, by designating the use or uses to be made of the water and by setting criteria necessary to protect the uses" (40 CFR § 131.2). Further, "such standards serve the dual purposes of establishing the water quality goals for a specific water body and serve

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[14] See U.S. EPA, 2005c
[15] See U.S. EPA, 1998
[16] See Krupnick et al., 1998
[17] See Chesapeake Bay Program, 2003a,b

as the regulatory basis for the establishment of water quality-based treatment controls and strategies beyond the technology-based levels of treatment required by sections 301(b) and 306 of the Act" (40 CFR § 131.2).

The EPA sets minimum requirements for approvable standards and criteria including: use designations; water quality criteria sufficient to protect the designated uses; and an antidigradation policy (40 CFR § 131.6). Traditionally, Nr and other land, air, and water pollutants are measured in terms of quantity (mass) released per unit time (e.g., kg/day) or as a concentration (e.g., milligrams per liter, ml/L). Therefore, regulations often specify mass loading limits or maximum concentrations in permits.

In the mid-to-late 1990s, EPA began to emphasize the development of numeric nutrient criteria for both P and N through the state standards-setting process because, according to the 1996 Water Quality Report to Congress (U.S.EPA, 1997), 40% of the rivers, 51% of the lakes and ponds, and 57% of the estuaries assessed for the report were exhibiting a nutrient-related impairment. Few states had adopted numeric nutrient criteria for all affected waterbodies, especially for N, often relying on narrative criteria or secondary effects such as chlorophyll-*a* concentration, dissolved O_2, or water clarity. EPA's strategy, driven by President Clinton's Clean Water Action Plan (U.S. EPA and USDA, 1998) mandated numeric nutrient criteria to begin to address the problem (U.S. EPA, 1999). To move the objectives of the Clean Water Action Plan forward, EPA published national nutrient criteria guidance for lakes and reservoirs (U.S. EPA, 2000d), rivers and streams (U.S. EPA, 2000b), estuaries and coastal waters (U.S. EPA, 2001b), and wetlands (U.S. EPA, 2007c), based upon ecoregional guidance for lakes and reservoirs and rivers and streams. To date, relatively few states have adopted new numeric criteria into their water quality standards. While some successes are evident in promulgating P criteria for freshwater systems, which has a richer history of numeric criteria incorporation into state water quality standards, development of numeric nitrogen criteria has been elusive for a variety of reasons.

Nr management in multiple media and across jurisdictions can be complicated because the CWA has little authority over atmospheric sources, and individual states explicitly lack authority to control upstream sources. For example, extensive monitoring and analysis of the sources of reactive nitrogen in the Raccoon River of western Iowa have shown that point sources from municipal treatment plants and residential septic tanks account for less than 8% of the total nitrogen load to the system, with agricultural runoff being the overwhelming source (Jha et al., 2010). This disparity is similar statewide (Libra et al., 2004). As a result, nutrient management strategies that are focused on the control of point sources can often result in inefficient allocation of resources if non-point sources are not also addressed. In addition it is often the case for estuaries such as the Gulf of Mexico or Chesapeake Bay, that management goals that meet water quality standards cannot be attained without interstate compacts or a strong federal role. This may be resisted by upstream states that may have to bear the cost but do not necessarily reap the benefits of the water quality improvement. Such a dilemma underscores the need for an integrated approach to Nr management. The Committee notes that a State-EPA Nutrient Innovations Task Group has considered some options for improving control of nutrient pollution sources (State-EPA Nutrient Innovations Task Group, 2009).

Populated (urban/suburban/developed) land areas provide significant loads of Nr to the environment, both by generation (e.g., deposition of NO_x emissions) and by transfer (e.g., domestic sewage from food imported into the watershed). Categorical sources include sewage treatment plants (STPs), industries, subsurface (septic) systems, atmospheric deposition, domestic animal and wildlife waste, and fertilizers used on lawns, gardens and landscapes. Infrastructure (e.g., storm sewers) and landscape conditions (e.g., increased impervious cover) more efficiently move Nr associated with surface runoff to receiving waters and may also inject or infiltrate Nr into ground water. Landscape changes, primarily increases in impervious cover, soil disturbance and compaction, and wetland/hydric soil losses, have also reduced the capacity for natural systems to treat Nr inputs by recycling or denitrification. Other disruptions in chemical condition (e.g., acidification), biology (e.g., vegetative cover), and physical character (e.g., temperature increase) alter the nitrogen cascade, which may have both negative and positive consequences for Nr amelioration on the populated landscape and in air and water. Populated lands are estimated to export as much as 10 times the total nitrogen that was exported under pre-development conditions.

Finding 15: Intervention to control Nr under most water management programs generally occurs in three ways:

- Prevention or source controls.
- Physical, chemical, or biological "dead ending" or storage within landscape compartments where it is rendered less harmful (e.g., long-term storage in soils or vegetation; denitrification, primarily in wetlands; reuse).
- Treatment using engineered systems such as wastewater treatment plants or BMPs for stormwater and nonpoint source runoff.

While most management programs focus on the third (treatment) approach, there are opportunities for combining the three that can be more effective and cost less. Furthermore, it is important to recognize that in some cases total reduction of water impacts of Nr may rely nearly as much on stricter enforcement of the Clean Air Act as the Clean Water Act.

Recommendation 15: *To better address Nr runoff and discharges from the peopled landscape the Committee recommends that EPA:*

15a.

- *Evaluate the suite of regulatory and non-regulatory tools used to manage Nr in populated areas from nonpoint sources, stormwater and domestic sewage, and industrial wastewater treatment facilities, including goal-setting through water quality standards and criteria.*
- *Determine the most effective regulatory and voluntary mechanisms to apply to each source type (recognizing that in some cases total reduction of the impacts of Nr may rely nearly as much on stricter enforcement of the Clean Air Act as the Clean Water Act) with special attention to the need to regulate nonpoint source and related land use practices.*

15b.

- *Review current regulatory practices for point sources, including both wastewater treatment plants and stormwater, to determine adequacy and capacity towards meeting national Nr management goals.*
- *Consider technology limitations, multiple pollutant benefits, and funding mechanisms as well as potential impacts on climate change from energy use and greenhouse gas emissions, including nitrous oxide.*

15c.

- *Set Nr management goals on a regional/local basis, as appropriate, to ensure most effective use of limited management dollars.*
- *Fully consider "green" management practices such as low- impact development and conservation measures that preserve or re-establish Nr-removing features to the landscape as part of an integrated management strategy, along with traditional engineered best management practices.*

15d.

- *Research best management practices that are effective in controlling Nr, especially for nonpoint and stormwater sources, including land and landscape feature preservation and set Nr management targets that realistically reflect these management and preservation capacities.*
- *Construct a decision framework to assess and determine implementation actions consistent with management goals.*

15e.

- *In cooperation with the Departments of Agriculture and Army, the Fish and Wildlife Service and the Federal Emergency Management Agency, the EPA should develop programs to encourage wetland restoration and creation with strategic placement of these wetlands where reactive nitrogen is highest in ditches, streams, and rivers. The Agency should also address the means of financing, governance, monitoring, and verification. Such programs might be modeled on the Conservation Reserve Program or extant water quality and environmental trading programs, but need not be limited to current practices (as discussed in section 5.3.4).*

4.4. Water Quality Monitoring and Assessment

Under Section 106 of the CWA, the EPA provides funds to assist state and interstate agencies and tribes to conduct monitoring of the nation's waters to ensure adopted water quality criteria and designated uses are met. Further, primarily under Section 305(b) of the CWA, those entities are required to report, on a biennial basis, on the health and status of their jurisdictional waters. These assessments are presented by the states to the EPA to categorize attainment of designated uses. EPA published these reports up until 1998 (U.S. EPA, 2000a), after which it transitioned into a Water Quality Report in 2000 (U.S. EPA, 2002) and a National Assessment Database in 2002 (U.S. EPA, 2010c). States also prepare a list of "impaired" waters under Section 303(d) of the CWA and EPA develops a synthesis of the CWA Section 305(b) and 303(d) reporting under a Consolidated Assessment and Listing Methodology (CALM) approach.

As discussed above, the EPA compiles the approved state 303(d) lists into a national listing (U.S. EPA, 2010e). The list provides information by state as well as by impairment cause, and identifies the TMDLs completed to date. The most current data available on the EPA Web site includes reporting from most entities through 2008. The report identifies 6,816 impairments related to "nutrients" (almost 9% of all identified impairments), although other impairments may ultimately have a nutrient enrichment cause. For example, organic enrichment/oxygen depletion (6,410), turbidity (3,046), noxious aquatic plants (981), algal growth (539), and ammonia (general toxicity 356), can all have a common cause such as N or P enrichment. It should also be clear that impairments may have multiple causes so, for example, waters identified as impaired by O_2 depletion may also be impaired by nutrients.

There are other initiatives promoted by EPA to monitor and assess the nation's waters, generally implemented in collaboration with, or by, the state and interstate agencies and tribes having jurisdiction over the waters. These include the Wadeable Stream Assessment (U.S. EPA, 2006c), the National Coastal Assessment and its National Coastal Condition Reports (U.S. EPA, 2001a, 2004a, 2006b), the Survey of the Nation's Lakes and Survey of the Nation's Rivers and Streams, and more recently, probabilistic monitoring efforts in lakes, streams, and estuaries (U.S. EPA, 2010d). Many of these are aimed at including a biological assessment component that is often lacking in water pollutant and chemistry efforts described above.

The USGS collects data on surface and underground waters and disseminates these data to the public, state and local governments, public and private utilities, and other federal agencies involved with managing water resources. The Committee encourages EPA to work closely with USGS on monitoring and assessment activities.

The National Oceanic and Atmospheric Administration has periodically produced estuarine assessments under the National Estuarine Eutrophication Assessment (NEEA) program. The most recent report was released in 2007 (Bricker et al., 2007). The report has a focus on nutrient enrichment and its manifestations in the estuarine environment and relies on participation and interviews of local experts to provide data for the assessment. Among the key findings were:

- Eutrophication is a widespread problem, with the majority of assessed estuaries showing signs of eutrophication – 65% of the assessed systems, representing 78% of assessed estuarine area, had moderate to high overall eutrophic conditions.
- The most common symptoms of eutrophication were high spatial coverage and frequency of elevated chlorophyll-*a* (phytoplankton) – 50% of the assessed estuaries, representing 72% of assessed area, had excessive chlorophyll-a ratings.

4.5. Clean Air Act and Air Quality Regulation and Management

The modern history of American air pollution control legislation begins with the 1963 Clean Air Act (CAA) which, along with its amendments, requires the EPA to establish and revise National Ambient Air Quality Standards (NAAQS) and to prepare state of the science reviews such as the Criteria Documents and more recently the Integrated Science Assessments (ISA) (U.S. EPA, 2005a, 2006a, 2007a). There are six criteria pollutants: carbon monoxide, lead, NO_2, ozone, SO_2, and PM. These have been determined to endanger public health or welfare. The CAA as currently written requires a review of the scientific criteria for these standards at five-year intervals. Although NO_2 is the only Nr compound specified as a criteria pollutant, NH_x and NO_y play a major role in formation of the secondary pollutants ozone and particulate matter.

The CAA has been amended several times since its inception. In 1970, the CAA was amended "to provide for a more effective program to improve the quality of the nation's air." The CAA was amended again in 1977, primarily to mandate reductions of emissions from automobiles. Despite evidence that NO_x is the central pollutant in photochemical smog formation (Chameides and Walker, 1973; Crutzen, 1973, 1974; Fishman and Crutzen, 1978; Fishman, et al., 1979), federal regulations did not require automobiles to control NO_x emissions to below 1 g/mi (0.14 g N per km) until 1981. Few locales violate the standards for NO_2,[18] but the secondary effects of several of these gases also pose health and welfare concerns. If a city had an annual average NO_2 level

Table 13: Federal primary ambient air quality standards that involve Nr, effective February 2010.

Pollutant	Federal Primary Standard (NAAQS)
Ozone (O_3) 1-hr average 8-hr average	 0.12 ppmv 0.08 ppmv
Nitrogen Dioxide (NO_2) 1-hr average Annual average	 100 ppb 0.053 ppmv (100 µg/m^3)
Particulate Matter, coarse (PM_{10}) Diameter ≤ 10 µm, 24-hr average Annual average **Particulate Matter, fine ($PM_{2.5}$)** Diameter ≤ 2.5 µm, 24-hr average Annual average	 150 µg/m^3 50 µg/m$^-$ 35 µg/m^3 15 µg/m^3

Note: Secondary standards are currently identical to the primary standards. Source: www.epa.gov/air/criteria.html

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[18] In 2010, EPA promulgated a new 1-hour standard of 100 ppb for NO_2 [Primary National Ambient Air Quality Standards for Nitrogen Dioxide; Final Rule, *Federal Register* 75 (26): 6474-6537]. Monitoring for compliance with this new standard is required, but it will not be known for several years which if any locales violate this standard.

anywhere near the NAAQS for NO_2, it would risk severe photochemical smog – the summertime efficiency for ozone production ranges from 4 to 10 ppb O_3 per ppb NO_x.

As previously discussed, the focus on compliance monitoring for NO_2 ignores the other, equally important members of the NO_y family such as HNO_3 that deposits quickly onto the earth's surface. It is clear that a causal relationship exists between current levels of N and S deposition and numerous biologically adverse effects on ecosystems across the U.S. (U.S. EPA, 2008d). Conversion of the existing network of NO_x monitors to NO_y monitors with a detection limit of 0.1 ppb would still demonstrate compliance with the NO_2 standard but greatly increase the utility of the measurements for model evaluation as well as for understanding nitrate deposition and formation of photochemical smog, and haze.

Air pollution, especially ozone and PM, continued to be a problem in many American cities and the CAA was again amended in 1990. The Nr-relevant aspects were aimed at controlling urban smog and acid deposition. States were required to develop emissions inventories for reactive organic compounds, carbon monoxide, and NO_x, but not NH_3 or N_2O. Over the U.S., sulfate and nitrate are responsible for about two-thirds and one-third, respectively, of the direct deposition of acids. The CAA Amendment of 1990 required emissions decreases of 10 million tons of SO_2 and 2 million tons of NO_x relative to 1980 levels. Ammonia and ammonium, although they contribute to acidity after entering terrestrial ecosystems (Galloway et al., 2003; NRC, 2003) and are expected to play an increasing role (Pinder et al., 2008), were not regulated by this legislation.

The 1997 revision of the CAA and related regulations changed the standards for ozone and PM (see Table 13). A sizable fraction of the mass of PM less than 2.5 microns, $PM_{2.5}$, is condensed Nr. As stated above, these particles have adverse health consequences. PM is also controlled by the Regional Haze Regulations (40 CFR 51). These regulations require that by the year 2064, states must restore Class I areas defined in the regulations to their natural levels of atmospheric clarity.

Ozone and PM, the two most recalcitrant of the criteria pollutants, cover large spatial scales. All of the ozone and much of PM are secondary pollutants in that they are not released at the tailpipe but form in the atmosphere. Ample evidence shows that much or most of the PM in American cities is secondary (e.g., Donahue et al., 2009). Violations are declared on urban scales, responsibility for their control was assigned to states, but the physics and chemistry of smog and haze are regional. In the eastern U.S., ozone episodes often cover several states and involve pollutants emitted in upwind states that do not themselves experience violations (Husar et al., 1977; Logan 1989; Moy et al., 1994; Ryan et al. 1998). The 1990 amendments to the Clean Air Act authorized, in part as a response to this scaling problem, the Ozone Transport Assessment Group (OTAG) and the Ozone Transport Commission (OTC). These have jurisdiction extending from Washington, D.C. to Maine. Progress has been made on regional control of emissions; the NO_x State Implementation Plan (SIP) call, implemented in 2003 and 2004, has led to measurable improvements in ambient ozone and nitrate levels (Gego et al., 2007; Sickles and Shadwick, 2007a). Experiences with ozone and PM provide a useful demonstration of why it is necessary to develop an integrated approach to management of Nr.

Atmospheric thresholds for Nr

As shown in Table 13 the metric used for safe, upper limits in the atmospheric environment is concentration (in mass per unit volume of air or volume mixing ratios) averaged for a given time period, usually 1 hour, 8 hours, 24 hours, or annually. The thresholds for excess Nr in the atmosphere remain an area of active research. The only Nr compound for which there is currently a NAAQS is NO_2, which may not exceed 0.053 ppm (100 $\mu g/m^3$) for the annual arithmetic mean and 100 ppb for the one-hour average. This standard, based on the direct health effects, is certainly inadequate because NO_2 concentrations well below 0.053 ppm lead to concentrations of secondary pollutants well above acceptable levels (i.e., $PM_{2.5}$ and O_3). The NO_2 concentration required to achieve the current 75 ppb ozone standard has not been rigorously established, but it must be well below 0.053 ppm, because information provided by EPA indicates that areas currently in violation of the ozone standard typically have NO_2 concentrations below 0.020 ppm (U.S.EPA, 2010a). The NO_2 concentration required to achieve the current 15 $\mu g/m^3$ $PM_{2.5}$ standard is probably also below the 100 $\mu g/m^3$ standard for NO_2 because of the role of NO_2 in secondary particulate formation. States in the eastern U.S. are considering substantial additional NO_x emissions reductions in order to comply with the new 8-hour 75 ppb ozone standard. One scenario being tested (G. Aburn, Maryland Department of Environment, personal communication) involves the following reductions: (1) reducing NO_x emissions for point sources by 65%, (2) reducing NO_x emissions for on-road sources by 75 percent, (3) reducing NO_x emissions for nonroad sources by 35%, and (4) reducing VOC emissions by 30% for all source groups.

As further discussed in Section 6.2, it is the opinion of the Committee that a decrease in NO_x emissions of 2 Tg N/yr relative to the 2002 baseline level can be achieved in the near term. Emissions decreases implemented since 2002 have already substantially improved ozone concentrations (Gégo et al., 2007). The absolute amount of decrease and the positive impact it would have on human health is region dependent, but further decreases will result in further beneficial decreases in $PM_{2.5}$ and O_3 concentrations.

The threshold for total Nr in the atmosphere is yet to be fixed, but depends on its rate of deposition to the surface and the sensitivity of the receptor(s). The immediate need for determining thresholds for atmospheric Nr is monitoring of NO_y and NH_x.

4.6. Thresholds for Excess Nr Effects on Terrestrial Ecosystems

In parallel with the original concept of critical loads developed by Nilsson and Grennfelt in 1988 and now widely used for air quality management in Europe (Appendix D), thresholds in general and critical loads specifically for Nr effects on terrestrial ecosystems in the U.S. should be understood to be "quantitative estimates of exposure to air concentrations of Nr compounds below which harmful effects on specified sensitive elements within ecosystem of concern do not occur according to present knowledge" (Nilsson and Grennfelt, 1988; Heittelingh et al., 2001).

In developing these quantitative estimates of thresholds and/or critical loads for terrestrial ecosystems in the U.S. (e.g., Fenn et al., 2003), it is imperative to understand the extraordinarily wide diversity and Nr-sensitivity of various components of terrestrial ecosystems in different parts of the U.S., as well as the huge differences in purposes and intensity of management and public perceptions of the value of these ecosystem components to various sectors of American society. Thus, the critical loads appropriate for maintaining species diversity in a natural grassland in northern Minnesota or a wilderness area in the Mediterranean climate of southern California are likely to be very different from those associated with direct effects on similar systems in other regions of the U.S. – or even for beneficial and/or adverse effects on other components of the same terrestrial ecosystem. For example, the threshold or critical load for adverse effects of excess Nr on understory vegetation, beneficial mycorrhizae, or lichen communities in a forest ecosystem is likely to be very different from the threshold for adverse effects on the dominant forest trees in that same ecosystem. Thus, public perceptions of "specified sensitive elements within the ecosystem" may be important in determining what specific thresholds or critical loads should be considered in order to minimize or avoid specific adverse effects of concern.

At present, the sum total of directly measured wet plus dry-deposited chemically oxidized (NO_y) and chemically reduced (NH_x) inorganic Nr loads in various states within the contiguous U.S. are on the order of 3 to 15 kg N/ha/year (NADP, 2010; CASTNET, 2010). As shown in Appendix A, a three-year run of the Community Multiscale Air Quality (CMAQ) model also provided estimates of the average annual total Nr loads (including organic forms as well as inorganic NO_y and NH_x forms of Nr) in the contiguous U.S. These model estimates varied from minimal deposition values of about 3 kg N/ha/year to maximum estimated values of about 17 kg N/ha/year. This range agrees well with the range of the measurements.

These directly measured and modeled estimates of total (wet plus dry) deposition of organic and inorganic forms of Nr indicate that there are several areas, especially in the eastern U.S., and a few areas of the western U.S., where current total Nr loads are already very close to, or will very likely soon exceed, the recommended threshold and critical load estimates provided by Bobbink et al. (2010) in their review of scientific evidence regarding the impacts of atmospheric nitrogen deposition on plant diversity in terrestrial ecosystems.

4.7. Comments on Nr Critical Loads

In recent years, the Acid Rain Action Plan developed by New England governors and eastern Canadian Premiers has led to evaluations of critical loads to surface waters and forests in that region. Those studies identified many waters and forest lands that met or exceeded critical load capacity for combined sulfur and nitrogen deposition both in the New England States and in the eastern Canadian provinces. The plan set target decreases of 20 to 30% for nitrogen oxide emissions by 2007 and a 50% decrease in sulfur dioxide emissions by 2010. These targets are intended to decrease long-range transport of air pollutants, acid deposition, and nutrient enrichment of marine waters in this region.

In May 2006, a Multi-Agency Critical Loads Workshop was held, which led to the formation of a Critical Loads Ad-Hoc Committee (CLAD) within the National Atmospheric Deposition Program (NADP). A goal of the program is to "provide consistency in development and use of critical loads in the U.S." One outcome is a project undertaken by the Northeast States for Coordinated Air Use Management (NESCAUM) to: "estimate critical loads of sulfur and nitrogen in atmospheric deposition for areas where sufficient knowledge, data, and methods exist" and "to demonstrate the use of critical loads as a tool for assessing environmental policies and programs and managing natural resources."

A February 2007 Workshop sponsored by EPA on "The Assessment of Health Science for the Review of the National Ambient Air Quality Standards (NAAQS) for Nitrogen (NO_x) and Sulfur Oxides (SO_x)" expansively reviewed both ecosystem and human health effects toward revision of the NAAQS. Policy discussions at this workshop raised the questions of whether critical loads assessments were an effective means of improving ecosystem management, and whether the science was understood well enough to use critical loads as a management tool. The conclusion was that, although there was a substantial body of accumulated scientific evidence, there was only limited use of critical loads approaches for management of air quality in the U.S. The Multi-Agency Workshop on Critical Loads (mentioned above) was cited at EPA's 2007 workshop as an agenda-setting effort to resolve some of the science and policy issues that could help advance critical loads approaches in the U.S. The Integrated Nitrogen Committee believes that the primary reason critical loads are not now used in the U.S. is that policy makers in this country have so far not been willing to adopt unfamiliar air and water quality management approaches or approaches that have not been evaluated directly in this country. Thus, the Committee recommends that EPA consider implementation of the critical loads concept for management of deleterious Nr effects in various parts of the U.S.

Finding 16: The Committee finds that there have been persistent increases in the amounts of Nr that have been emitted into and retained within various ecosystems, affecting their functioning. Unless this trend is reversed, it will become increasingly difficult for many of these ecosystems to provide the services upon which human well-being is dependent. The Committee believes that there is a need to regulate certain forms of Nr to address specific problems related to excess Nr, and we believe that the best approach for an overall management strategy is the concept of defining acceptable total Nr critical loads for a given environmental system.

Recommendation 16: *The Committee recommends that the Agency work toward adopting the critical loads approach concept in determining thresholds for effects of excess Nr on terrestrial and aquatic ecosystems. In carrying out this recommendation the Committee recognizes that it will in many cases be necessary for the Agency to enter into new types of research, policy, and regulatory agreements with other federal, state, and tribal units based on cooperative, adaptive, and systemic approaches that derive from a common understanding of the nitrogen cascade.*

4.8. Tradeoffs of Nr Impacts in Risk Reduction Strategies

Because nitrogen is such an abundant and widespread element, and Nr is such a critical component of the earth's biosphere, associated impacts are many and pervasive. In many cases, strategies to manage the impacts of Nr involve tradeoffs, i.e., mitigating one type of impact may exacerbate others. Given the interactions among oxidized and reduced N species, it is important to recognize the potential for unintended consequences to occur as a result of strategies that are aimed at limiting one form of Nr in air or water but lead to the increased production of other forms of Nr, or the formation and release of other contaminants of concern. For example, stringent control of point sources of Nr can be energy-intensive, requiring significant energy investments for chemicals, electricity, and other support, and this may in turn lead to the production of more reactive nitrogen and increased CO_2 emissions. Furthermore, there may be environmental impacts of these treatment processes, particularly in the production of solid wastes that can be significant environmental hazards. This is the main reason that a life cycle approach is necessary in evaluating any remediation or treatment scheme. In addition, as discussed in Section 3.1.2, numerous lakes, reservoirs, rivers, and fjords worldwide exhibit N and P co-limitation, either simultaneously or in seasonally-shifting patterns. Therefore, strategies are needed to reduce both P and N inputs. Not all control practices will be effective for dual nutrient reduction and this must be taken into consideration. Four categories of tradeoffs examined below are: ammonia release from concentrated feed lot operations (CAFOs), concerns about human nutrition, nitrification and denitrification, and nitrogen-carbon related impacts.

Ammonia release from CAFOs

As a result of effluent guidelines for NH_3 in aquatic systems, state and federal regulations and programs under the CWA were developed to address water quality protection from CAFOs. The resulting manure management systems utilized NH_3 volatilization as a means to remove N and decrease the N in the manure when land applied. Only recently has the resulting increase in NH_3 emission into the air been viewed as a potential problem with respect to air quality concerns and N deposition.

Finding 17: Current EPA policy (40 CFR Part 51, Clean Air Fine Particle Implementation Rule) discourages states from controlling ammonia emissions as part of their plan for reducing $PM_{2.5}$ concentrations. In this rulemaking, EPA states that "ammonia reductions may be effective and appropriate for reducing $PM_{2.5}$ concentrations in selected locations, but in other locations such reductions may lead to minimal reductions in $PM_{2.5}$ concentrations and increased atmospheric acidity." Ammonia is a substantial component of $PM_{2.5}$ in most polluted areas of the United States at most times. While it is true that reducing NH_3 emissions might increase the acidity of aerosols and precipitation, the net effect of NH_3 on aquatic and terrestrial ecosystems is to increase acidity. After being deposited onto the earth's surface, NH_4^+ is under most circumstances quickly nitrified, increasing the acidity of soils and waters. The Committee is unaware of any evidence that NH_3 reduces the toxicity of atmospheric aerosols or that high concentrations of NH_3 occur naturally over any substantive area of the United States. It has not yet been established which components of PM have substantive impacts on human health, but the total concentration of $PM_{2.5}$ correlates with morbidity and mortality, and NH_3 contributes to $PM_{2.5}$. The visibility degradation and other adverse effects associated with $PM_{2.5}$ are related to aerosol surface area or mass where NH_4^+ certainly plays a role.

Recommendation 17: *The Committee recommends that the EPA presumption that NH_3 is not a $PM_{2.5}$ precursor should be reversed and states should be encouraged to address NH_3 as a harmful $PM_{2.5}$ precursor.*

Swapping N between environmental systems

Nitrous oxide is produced in "natural" and agricultural soils, and all aquatic systems, almost exclusively as a result of the microbial processes of nitrification and denitrification. As NH_4^+ ion is the initial mineral N product formed during organic matter mineralization and most of the fertilizer used worldwide is NH_4^+ based (e.g., urea, ammonium sulfate) (FAO, 2007), the suite of microbiological reactions that result in the release of gaseous N products need to be considered.

Nitrification is the oxidation of NH_4^+ ion to NO_3^- (Figure 17). Most commonly, nitrification is a chemolithotropic process consisting of the conversion of ammonium to nitrite, which is then converted to NO_3^- by a second group of bacteria. The ammonium oxidizing bacteria (AOB) are obligate aerobes with some species that are tolerant of low-oxygen environments. The most common genera of autotrophic NH_4^+ oxidizers are *Nitrosospira* and *Nitrosomonas*. AOB are found in most aerobic environments where ammonium is available through the mineralization of organic matter or where N compounds are added.

Biological denitrification is the dissimilatory reduction of NO_3^- and nitrite to produce NO, N_2O, and N_2 by a taxonomically diverse group of bacteria. These bacteria synthesize a series of reductases that enable them to utilize successively more reduced N oxides as electron acceptors in the absence of oxygen. The general reductive sequence is shown in Figure 17. In addition to the free-living denitrifiers, symbiotically-living Rhizobia in root nodules of legumes are able to denitrify nitrate and produce nitrous oxide (Mosier and Parkin, 2007).

The abundant denitrifiers are heterotrophs, which require sources of electron-reducing equivalents contained in available organic matter. Factors that most strongly influence denitrification are oxygen, nitrate concentration, pH, temperature, and organic carbon. The reductive enzymes are repressed by oxygen but not by NH_4^+. Nitrous oxide reductase appears to be more sensitive to oxygen than either NO_3^- or nitrite reductase. Therefore N_2 production predominates in more anoxic sites and N_2O production may be greater in more aerobic conditions. However, the ratio of N_2 to N_2O emitted may also be affected by high NO_3^- concentrations and associated higher levels of electrical conductivity and osmotic stress and soil pH (low pH favors N_2O production).

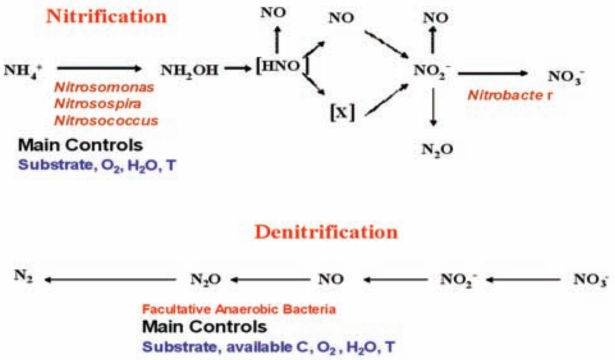

Figure 17: Diagram of the nitrification and denitrification processes

Source: Mosier and Parkin, 2007. Reprinted with permission; copyright 2007, Taylor & Francis Group LLC – Books.

Given these interactions among oxidized and reduced N species (discussed above), it is important to recognize the potential for unintended consequences to occur as a result of strategies that may be aimed at limiting one form of Nr in air or water but lead to the increased production of other forms of Nr. One such instance is the potential offsetting of the benefits of NO_3^- remediation at the expense of increasing input of N_2O to the atmosphere. An example of such a situation involves NO_3^- leached from agricultural fields, much of which could be removed from drainage water in natural or reconstructed wetlands. This process is ideal if the denitrification process goes to completion, i.e., only N_2 is produced. If, however, the process is incomplete, and NO and N_2O gases are emitted, then the end result may create a compensating risk that could be greater than that posed by the nitrate that is removed. This is because NO continues to be reactive in the atmosphere and is eventually redeposited in aquatic or terrestrial systems, and N_2O is a GHG that has an atmospheric life time of approximately 100 years and a radiative forcing of approximately 300 times that of CO_2 on a hundred-year time frame (IPCC, 2001). N_2O is also a major source of NO in the stratosphere and depletes stratospheric ozone (Crutzen, 1981). If more of the NO_3^- denitrified is converted to N_2O in wetlands than upstream or downstream, the environmental cost may be high. Hernandez and Mitsch (2007) found that permanently flooded wetlands had lower N_2O/N_2 ratios of emissions than did intermittently flooded wetlands. They also found that the ratio was higher in the cold months even though the flux rates are much lower then. A full risk assessment needs to be made to determine how much of such "pollutant swapping" is advisable.

A similar potential exists for Nr mediation in sewage treatment. The current practice is to convert ammonia/ammonium that mineralizes from excreted organic matter to nitrate through the nitrification process. As nitrate-containing effluent from sewage treatment flows into aquatic systems the nitrate may be denitrified, resulting in N_2O production if denitrification is not complete. The protein consumption by some 301 million humans in the U.S. results in the processing of ~ 2 Tg of N annually (~18.4 g N/ person/day), much of which flows through sewage treatment facilities and ultimately leads to the production of between 0.06 and 0.1 Tg of N_2O-N /yr in aquatic systems or soils to which sewage sludge is applied.

Tradeoffs among C and N-driven impacts

Reactive N also contributes to many impacts on the environment that are also impacted by other chemical species, notably carbon. As depicted in Figure 18, there are several points of tangency between the global C and N cycles. These are: combustion, agricultural production, industrial production, soil and sediment processes, and end-of-life disposition of products. The implication of these interactions is that, in many instances, the perturbation of one cycle cannot be fully assessed without including effects on the other. For example, proposals to develop bio-based products (biofuels, but also other products) as the preferable alternative to fossil-based resources are not free from impacts. Such trade-offs may involve a single impact (e.g., global climate change to which both carbonaceous gases and N_2O contribute) but may also involve trade-offs between impacts that are not easily compared. Figure 19 shows the latter case in the form of climate change impacts (to which C is a principal contributor) versus eutrophication impacts (to which nitrogen is a principal contributor) for several different biofeedstock-product combinations which are evaluated relative to the substituted commercial product made from fossil C. A value of 100% on the y-axis would mean that the bio-based alternative is no better than the fossil-based counter-product, while the negative region of the y-axis in Figure 19 represents net C sequestration. It is difficult to make direct comparisons across disparate impact categories, however Figure 19 suggests that, in choosing among alternatives, policies that aim to minimize both sets of impacts would be preferred.

Finding 18: The Committee notes that the effective management of Nr in the environment must recognize the existence of tradeoffs across impact categories involving Nr transformations and the cycling of other elements.

Recommendation 18: *The Committee recommends that the integrated strategies for Nr management outlined in this report be developed in cognizance of the tradeoffs associated with reactive nitrogen in the environment (consistent with the systems approach of overarching recommendations 2 and 3 discussed in Section 6.2 of this report). Specific actions should include:*

- *Establishing a framework for the integrated management of carbon and reactive nitrogen;*
- *Implementing a research program that addresses the impacts of tradeoffs associated with management strategies for carbon, reactive nitrogen, phosphorus, and other contaminants of concern;*
- *Implementing a research and monitoring program aimed at developing an understanding of the combined impacts of different nitrogen management strategies on the interchange of reactive nitrogen across environmental media.*

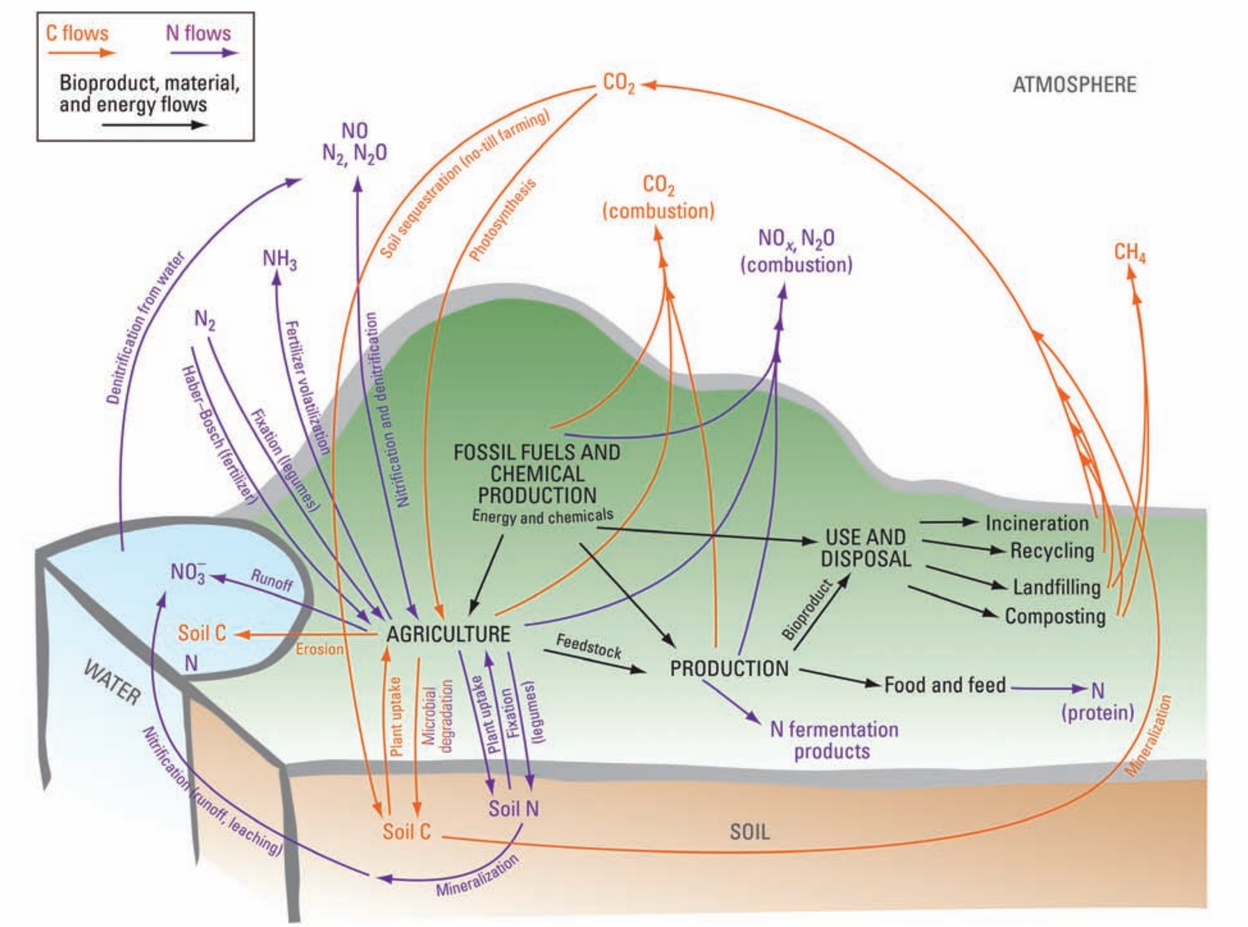

Figure 18: Combined carbon and nitrogen global cycles

Source: Miller et al., 2007 (Figure 1, p. 5178). Reprinted with permission; copyright 2007, American Chemical Society.

4.9. Interactions of the N Cascade and Climate

Weather and climate vary substantially on many time scales including the interannual. Long-term (decadal or more) changes in climate as have been predicted by IPCC (2007a,b) may have profound effects on the N cycle; conversely, changes in the biogeochemical cycle of Nr can induce climate forcing. While it is beyond the scope of this report to fully address how cycles of C and N interact (see Figure 18 for a general treatment of the intersection points of C and N cycles), there are several ways in which climate impacts the biogeochemical cycle of Nr and vice versa (e.g., Yienger and Levy, 1995; Holland et al., 1997; Hungate et al., 2003; Hungate et al., 2004; Sutton et al., 2007; Thornton et al., 2007; Levy et al., 2008; Sokolov et al., 2008). These are highly interactive and nonlinear systems. The following important interactions are noted:

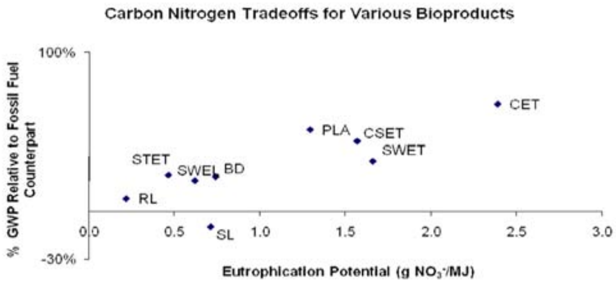

Figure 19: Comparisons between Global Warming Potential (GWP) and eutrophication impact categories for various bioproducts

Abbreviations: BD=Biodiesel; CET=Corn Ethanol; CSET=Corn & Stover Ethanol; PLA=Polylactic Acid (Corn); RL=Rapeseed Lubricant; SL=Soybean Lubricant; STET=Stover ethanol; SWEL=Switchgrass Electricity; SWET=Switchgrass Ethanol.

Source: Adapted from Miller et al., 2007 (Figure 2, p. 5180). Adapted with permission; copyright 2007, American Chemical Society.

- Increased deposition of Nr into terrestrial and aquatic ecosystems can alter the sequestration of carbon, while increased ambient CO_2 can change the deposition and uptake of Nr.
- Nitrate flux from fields to surface waters increases with increasing rainfall (see Box 5:The Impact of Climate Change on Agricultural Discharge of Reactive Nitrogen).
- Increasing temperature can both increase and decrease atmospheric loading of particulate matter.
- Aerosols (PM) have direct and indirect (through cloud microphysics) effects on radiative forcing of climate and on the hydrological cycle.
- N_2O and O_3 are greenhouse gases.
- Soil Nr chemistry and emissions of N_2O, NH_3, and NO depend on environmental conditions such as temperature and soil moisture.
- The amount of Nr deposited and exported from the U.S. depends on meteorological variables including wind speeds and convection.

Numerical models, when verified against past climates, can provide insight into possible future climates and their impacts on the nitrogen cycle. For example, increasing temperatures increase the amount of NO_x control necessary to achieve the same amount of photochemical smog control (Bloomer et al., 2009; Jacob and Winner, 2009). The EPA program for studying the impact of climate change on photochemical smog (air pollution ozone) production offers a useful model; see Jacob and Winner (2009) for an overview.

Finding 19: The biogeochemical cycle of Nr is linked to climate in profound, but nonlinear ways that are, at present, difficult to predict. Nevertheless, the potential for significant amplification of Nr-related impacts is substantial, and should be examined in more complete detail.

Recommendation 19: *The EPA should support cross-disciplinary and multiagency research on the interactions of climate and Nr. To determine the interactions of global biogeochemical Nr cycles and climate, the Committee suggests that EPA follow a series of steps such as:*

1. *Select several likely scenarios for global climate from the IPCC report for the year 2050.*
2. *Down-scale statistics or nest regional climate models within each of these global scenarios to generate meteorological and chemical fields (e.g., temperature, relative humidity, winds, precipitation, CO_2) for a few years around 2050.*
3. *Run several independent biogeochemical Nr models (earth system models that include air/water/land) for North America for these years with current Nr and emissions and application rates.*
4. *Rerun models with decreased Nr emissions/application to evaluate strategies for controlling impacts such as those described in this report.*

Box 3: The Impact of Climate Change on Agricultural Discharge of Reactive Nitrogen

The discharge of reactive nitrogen from intensively managed agroecosystems is characterized by a number of attributes that often exhibit a high degree of variability: fluctuating material flows associated with the degree of nitrogen fixation and the extent of denitrification, the interdependence of crops in rotation, and dependence on geography, weather patterns (particularly rainfall intensity, duration, and frequency), soil type, and agricultural practices.

One way to gauge the impact of climate change on such systems is to examine the ranges exhibited by historical data that collectively encompass the range of impacts that are anticipated. The assumption is that a changing climate will systematically alter governing attributes in plausibly predictable ways, for example increased annual rainfall and temperature over a large geographic region. The IPCC has provided general climate-induced impacts for world regions (IPCC, 2007a,b).

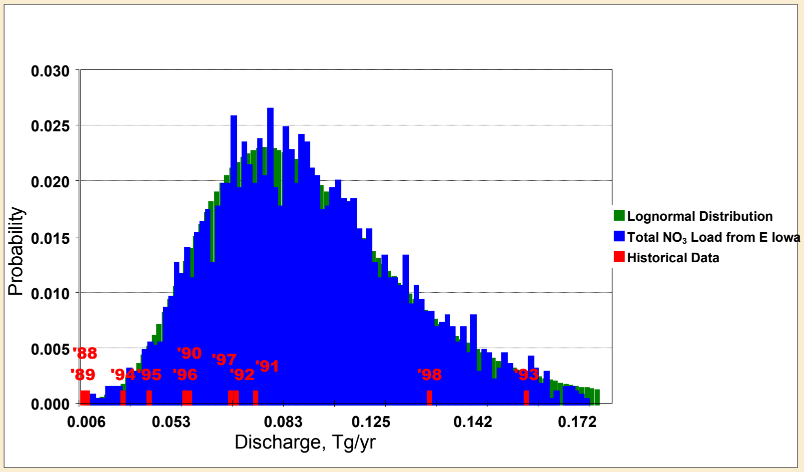

Figure 20: Probability of given discharge level for nitrate in the watersheds of eastern Iowa

Based on the simulation model of Miller et al. (2006). Red markers are historical data of discharges according to year as reported by Powers (2007). Green bars represent a log-normal distribution.

The general impact of climate change on the discharge of reactive nitrogen from agroecosystems can be discerned from the information in Figure 20. This figure shows a probability distribution for nitrate discharged from the watersheds of eastern Iowa (approximately 50,000 square km), which are dominated by corn-soybean agroecosystems (a general description of the region can be found in Kalkhoff et al., 2000). It is derived from information on the input of synthetic fertilizers in the region during the period 1989-1999, and includes factors that describe the transformation and transfer of Nr once applied. The distribution shown was generated using a Monte Carlo technique, details of which can be found in Miller et al. (2006). Also included in Figure 20 (in green) is a standard log-normal distribution, which the simulation most closely fits, and independently measured annual nitrate runoff data (an output of the system) over the same time period, as reported by Powers (2007). The simulation is not perfect, but it does capture the extremes of reactive nitrogen discharge, as represented by data for the years 1993 and 1998.

Figure 20 shows that the interannual variation in nitrate discharged is nearly 30-fold during the 11-year observation period. While the impact of climate change on such a system cannot be predicted for a given year, Figure 20 provides a basis for visualizing shifts in nitrate discharge due to changes in those factors that affect Nr transformation and transfer. For example a climate change scenario that predicts a general increase in precipitation amount and frequency, other factors being constant, will tend to shift the distribution of Figure 20 to the right, resulting in generally higher discharges of nitrate (see for example Vanni et al., 2001; the data point for 1993 in Figure 20 corresponds to precipitation in the region that was approximately 1.8 times the long-term annual average). Other factors, of course, may amplify or retard such impacts. Understanding whether or not implementation of best management practices and advanced technological methods can counteract climate change trends that favor increases in discharge would require a series of significant research studies and advances in modeling capabilities.

5

Integrated Risk Reduction Strategies for Nr

5.1. Importance of Integrated Risk Reduction Strategies

Chapters 3 and 4 of this report presented the environmental impacts and metrics associated with the emission of the various forms of Nr and reviewed ways of organizing these into impact "categories." As noted, Nr has many impacts on the environment, impacts that are interrelated through the nitrogen cascade. As previously stated, the nature of reactive nitrogen demands an integrated approach within EPA and across other relevant federal agencies, as reactive nitrogen cycles through the environment in different forms. A number of risk reduction approaches and the importance of considering Nr control points in the nitrogen cascade are discussed below.

5.2. Control Strategies for Nr

There are several ways in which the release and control of Nr in the environment can be approached. In general these can be classified as follows:

1. Improved practices and conservation – in which the flux of Nr that creates an impact is lowered through better management practices, including those that preserve or enhance Nr controlling ecosystem services (e.g., on-field agricultural practices, controlled combustion conditions, ecosystem function preservation and management)
2. Product substitution – in which a product is developed or promoted which has a lower dependency on or releases less Nr (e.g., N-bearing wastes instead of corn grain as a feedstock for biofuels, development of alternative power sources such as wind and solar)
3. Transformation – in which one form of nitrogen is converted to another form (e.g., nitrification of wastewater, denitrification in engineered or natural systems)
4. Source limitation – in which the amount of Nr introduced into the environment is lowered through preventive measures (e.g., controls on NO_x generation)
5. Removal – in which Nr is sequestered from impacting a particular resource (e.g., ion exchange)
6. Improved use or reuse efficiency – in which the efficiency of production that is dependent on Nr is improved (e.g., increased grain yields for lower Nr applied), or Nr wasted from one source is reused in another (e.g., algal farming)

Effective management of Nr requires combinations of these approaches; none is a perfect alternative for controlling Nr in the environment. Table 14 provides a summary of the pros and cons of each of these approaches.

5.3. Management Strategies for Nr in the Environment

Four types of management strategies for the control of Nr, and other pollutants, in the environment have evolved over the past 40 years:

1. Command-and-control – in which an entity's discharge of pollutants is regulated through a series of permitted limitations on emissions, violations of which may result in penalties being assessed
2. Government-based programs for effecting a policy, such as directed taxes, price supports for a given commodity, subsidies to bring about a particular end, and grants for capital expansion or improvement
3. Market-based instruments for pollution control in which market trading schemes are used to bring about a desired policy end, often at reduced overall cost.
4. Voluntary programs in which desired ends are achieved using private or government-initiated agreements or through outreach and education.

5.3.1. Command-and-control[19]

Policy makers have traditionally used command-and-control strategies requiring individuals and dischargers to meet mandatory guidelines. Such an approach evolved as the country was gearing up to meet the requirements first established nationally through the CWA and CAA enabling legislation in the 1970s. Because U.S. capabilities to monitor contaminant concentrations and predict environmental impacts were, generally, rudimentary, early emphasis was placed on "technology-based" approaches for managing emissions. This resulted in the promulgation of "best practicable technology" controls, and eventually "best available technology" controls, the idea being that mandating some level of control, even with uncertain improvements on impacts, would be better, and less arbitrary, than other approaches of the time.

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[19] Based on *Models in Environmental Regulatory Decision-Making,* National Research Council, 2007.

Table 14 : Advantages and limitations of various approaches to Nr control in forestry and agriculture

Control strategy	Advantages	Limitations
Improved practices, conservation	Lessens one or more impacts; utilization of existing ecosystem services	Education cost; availability and cost of preserved lands
Product substitution	Lessens the need for Nr, allows for more targeted uses of Nr	Questions of acceptability, technological issues
Transformation	Reduces one or more impacts to which Nr contributes, for denitrification closes the nitrogen cycle; utilizes natural biogeochemical processes that may be available ecosystem services	May contribute to other impacts; human presence has modified and diminished ecosystem service values
Source limitation	Reduces one or more impacts to which Nr contributes	Decreased crop yields, in some cases few viable alternatives yet developed
Removal	Reduces one or more impacts to which Nr contributes; natural land features/processes and ecosystem services may be used	Residuals containing Nr must still be managed effectively; availability, location and cost of land for natural or enhanced Nr removal
Improved efficiency	Reduces the need for Nr	Research and education costs

Nevertheless, both the CWA and the CAA had more specific goals that were aimed at protecting human health, public welfare, and ecosystem health. For example, the CAA required states to develop implementation plans (SIPs), the approval of which depended on their ability, once implemented, to meet ambient clean air standards. Likewise, the CWA required greater controls to be implemented for certain water bodies for which technology-based limits alone were insufficient to meet standards (this became the TMDL program).

Over time, and as our abilities to monitor, predict, and understand impacts improved, it became possible, or at least plausible, to tailor emission levels on a source-by-source basis, allowing the firm in question to decide its own technological approach. Thus permits, which place strict limits on the amount of pollution a firm is allowed to discharge over a specified period of time, have become the main method for managing the majority of point source contaminants, including the various forms of Nr, in the environment.

While the CWA has had considerable success in controlling point source discharges, it has been largely unsuccessful in limiting nonpoint discharges, and it is these sources that are particularly important for managing nutrient flows into receiving waters. The National Research Council has addressed this deficiency and pointed to the need to fully implement TMDL plans and establish numerical nutrient standards for nutrients (National Research Council, 2008b, 2009).

5.3.2. Government Taxes and Subsidies to Achieve Policy Ends

Government taxes and subsidies have created a variety of results, some in conflict with and some to further the ends of Nr management. Examples include U.S. agricultural and land-use policies, energy and transportation policies, and both point and nonpoint source mandated controls on N-bearing aquatic resources, including domestic and industrial wastewaters and agricultural runoff.

Current and future energy policy with respect to vehicle efficiency and biofuels will help determine the amount of Nr released into the environment from these sources. Some states have chosen to place modest taxes on fertilizer containing Nr, though the demand impact is slight at best. However, revenues may be dedicated to improved Nr utilization efficiency. Crop subsidies and crop insurance may at times expand land use and even encourage increased use of fertilizers, effectively increasing Nr in the environment. There are various agricultural conservation programs in the U.S. administered by the USDA. These include the Conservation Reserve Program and the Wetland Reserve Program (CRP and WRP). The former takes less suitable land out of cultivation and the latter encourages wetland protection and restoration. Both can contribute to better Nr management. The Environmental Quality Incentives Program (EQIP) directly subsidizes nutrient management efforts by crop and livestock producers. Of concern to the Committee is the need for more effective approaches aimed at encouraging farmers and land managers to adopt proven conservation and Nr management practices in fields and feedlots. The extent of proven practices, such as variable rate fertilizer application and installation of stream buffers, fall far below today's technological frontier.

5.3.3. Market-Based Instruments for Pollution Control[20]

A fundamental shift in environmental management philosophy was initiated with the 1990 Clean Air Act Amendments, which combined regulatory requirements with market flexibility allowing lower compliance costs through tradable credits. Most market-based policy instruments operate on the principle that if the regulatory framework or some other factor sufficiently alters the relative value of available decision choices for an individual or firm, subsequent decisions they make will be in alignment with the policy makers' objective.

As an example, if a government wants to limit pollution in a river where a number of polluters discharge, it need not adopt a uniform command and control limit on each discharger. Instead, a regulatory cap on the total pollutant loadings can be established and individual permit limits can be issued to all dischargers, with provisions that allow the dischargers to trade between their individual limits as long as the overall cap is not exceeded. Those dischargers having low pollution control costs will have incentive to control more pollution than their permit limit and thus generate water quality credits that can be sold to dischargers with high costs of pollution control. Because the overall cap on the pollutant is fixed, the regulatory goal is achieved. Water quality trading thus brings about the desired reduction in pollution level at lower cost than if all dischargers were required to use traditional onsite treatment technology. Water quality trading also encourages cost-effective pollution control investment by giving each firm a clear economic signal to invest in new technology to reduce pollution at a level that corresponds to the market value of the permit.

As with control strategies for Nr, there is no one universal market-based strategy that is applicable to every policy maker's objective. For example, the nature of incentives available to and effective with producers involved in over-fishing is different from landowners providing environmental amenities. In the former case, the objective is to restrict the intensity of fishing. In the latter case the objective is to encourage private landowners to provide environmental goods and services at the lowest cost possible.

Evolution of new market-based strategies is a continuous process. Most strategies have been customized over time to meet local needs. One can group such market based approaches under the following conceptual headings:

1. Water Quality Tradable Credits – Every polluting entity is allowed to discharge pollutants up to a certain pre-determined limit, defined in concordance with the terms of the CWA. The entities discharging less than their allocated limit generate credits. Under this strategy, credits can be traded with other polluting entities that have exceeded their allocated limit provided that water quality standards are not exceeded.
2. Auction-Based Contracting – Environmental or conservation contracts are auctioned, where individual landowners place their bids to provide such goods or services from their land. Two factors jointly determine the selection of the bids: the amount of the bid and the expected value of the environmental or conservation benefit resulting from accepting the bid.
3. Individual Transferable Quotas – An individual transferable quota (ITQ) is an allocation privilege to extract a specified quantity of a resource among a selected number of quota holders. The distinctive feature of the ITQ is that the privilege is transferable or leasable. An ITQ may be a right to produce under favorable circumstances, such as a tobacco quota when tobacco production would normally be limited.
4. Risk Indemnification for Specified Behavior – An example of this is crop insurance designed to protect farmers from uncertainty in the adoption of best management practices that provide a public good but are inherently riskier.
5. Easements – Conservation easements or conservation servitudes refer to the case in which a landowner enters into a legally binding agreement to surrender certain property rights for a specified period of time, either voluntarily or for compensation. Such arrangements usually provide public goods relative to the environment or conservation.

Policy maker objective, local conditions, and several other factors determine the suitability of a particular market based strategy. For example, water quality trading is well suited where there are a variety of dischargers at different levels of contribution and with varying control costs. A policy framework that facilitates the emergence of multiple options for dischargers to meet their permit limits, such as buying from more efficient controllers of discharge or investing in new equipment to achieve further reductions, is likely to accomplish the desired level of water quality at the least possible cost to the economy. Table 15 illustrates the potential effective application of a number of market-based approaches in specific situations. Accompanying this chapter are two examples of the application of

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[20] Based on Canchi, D., P. Bala and O. Doering, *Market Based Policy Instruments in Natural Resource Conservation,* Report for the Resource Economics and Social Sciences Division, NRCS, USDA, Washington D.C., March 3, 2006, pp. 4-9.

market-based approaches for the design of water quality trading schemes for Nr in watersheds (Box 4: Water Quality Trading to Meet the Long Island Sound Wasteload Allocation in Connecticut, and Appendix C: Water Quality Trading in the Illinois River Basin).

Table 15 shows pair-wise comparison between different market-based strategies. The objective and the incentive structure of the participants determine the suitability of one market based strategy over another. Each pair of cells briefly lists the most relevant set of conditions for which the respective strategy may be optimal (left cell points to strategy at the top of the column and right cell points to the strategy at the end of the row). Consider the two strategies (illustrated below): Auction-Based Contracting and Tradable Credit. If the participation of every private entity is essential, then Auction-Based Contracting works best. For example, if the objective is to preserve a large tract of privately owned contiguous land, Auction-Based Contracting is the appropriate strategy. This requires the participation of every private land owner to set aside a portion of their land. An auction designed to reveal the individual's land owner's reserve price for participation leads to the most efficient solution. A classic example of this is the Australian Government of Victoria's auction based Bush Tender program. Here it was essential to enlist blocks of land with particular hydrological and other characteristics to maximize the reduction in salinity and provide other environmental benefits. (Department of Sustainability and Environment, 2008). The goal was to bring about the reintroduction of native vegetation, its protection, and management where it would slow the development of salinity in soils. Offsets would not accomplish this, and as one looks at Table 15's characteristics of alternative approaches, neither would quotas, or insurance for BMPs. Easements might be used, but auctions were much more cost effective and more suited to the long term management commitment needed, as indicated in the second row of Table 15 under "Easements." Compared to this, if the objective is an overall reduction of a pollutant regardless of the individual private entity's contribution to the abatement, the Tradable Credit strategy with a cap is more appropriate. As another example, if aggregate depletion is of concern (as with fisheries) then individual transferable quotas are appropriate. However, auction-based contracting is preferable to individual quotas when no offsets exist.

Although there are significant differences between water and air quality trading, there are also several potential barriers to effective trading systems for both media. These are related to: accountability and monitoring; establishing standards and management goals; complexities of cross media and multiple source trading, including parity of sources; insurance that outcomes would reduce risk (environmental benefit); economics and marketability of traded credits; and transparency of the program, including public outreach and stakeholder involvement.

5.3.4. Biophysical and Technical Controls (control points) on Transfer and Transformations of Nr in and between Environmental Systems

Within the nitrogen cascade there are a number of places where the flow of Nr is constrained or regulated, either by nature or by human intervention, or a combination of the two. This report refers to these places in the cascade as "control" points. The control points may restrict the flow of Nr species within environmental systems (atmospheric, terrestrial, aquatic) or between them. The control points vary from primary controls where Nr is minimized through conservation measures or through after-the-fact measures that attempt to convert Nr that is emitted or not fully used to nonpolluting products (such as conversion to N_2 by denitrification or through long-term storage). The discussion of control points in this section is primarily focused on biophysical controls in terrestrial and aquatic environmental systems. However, the section concludes with a discussion of possibilities for decreasing NO_x emissions from combustion.

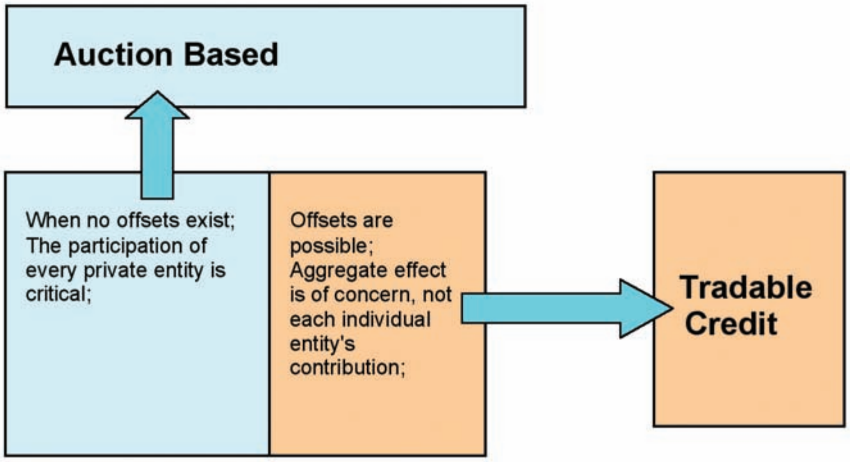

Table 15: Summary of market-based instruments for pollution control with conceptual examples

Auction Based Contracting		Individual Transferable Quotas		Insurance for the Adoption of BMPs		Easements		
When there exist no offsets; The participation of every private entity is critical;	Offsets are possible; Aggregate effect is of concern, not each individual entity's contribution;	When the depletion is of concern;	When the discharge is of concern;	Homogenous polluters; Offsets not feasible; Excessive pollution is primarily to mitigate uncertain profits; Modest short-term objective;	Not homogenous polluters; Offsets are possible; Pollution is an absolute consequence of the production process;	Unidirectional; When offsets are not possible; One entity retiring more property rights cannot trade with the other retiring less property rights.	Bidirectional; Offsets are possible; Requires specific action on the part of the participant to accomplish the objective;	**Tradable Credit**
		Aggregate depletion is of concern;	When there exist no offsets; The participation of every private entity is critical;	Tied to a production process; When risk averseness of the entity can be used to motivate participation;	Not tied to any production process; Suited for motivating participants to engage in secondary activities;	Auction based contracting can be seen as a refined and improved cost-efficient alternative to easements;	Designing of auction based contracting requires considerable professional expertise;	**Auction Based Contracting**
				Discharge of effluents is of concern;	Depletion of a resource is of concern;	Retirement of rights is of concern;	Acquisition of rights is of concern;	**Individual Transferable Quotas**
						No uncertainty; No action required on the part of the participant;	Tied to a production process:	**Insurance for the Adoption of BMPs**

Each pair of cells briefly lists the most relevant set of conditions for which the respective strategy may be optimal (left cell points to strategy at the top of the column and right cell points to the strategy at the end of the row).

Box 4: Water Quality Trading to Meet the Long Island Sound Wasteload Allocation in Connecticut

Pollutant trading is increasingly being promoted as a cost-effective means for attaining water quality standards. Connecticut and New York have been working with the EPA Long Island Sound Study (LISS) for more than 20 years to address low oxygen conditions (hypoxia) in Long Island Sound that have been linked to excessive loadings of nitrogen. A Total Maximum Daily Load (TMDL) for nitrogen, drafted by the two states and approved by the EPA in 2001, set a 58.5% nitrogen reduction target in 2014 from point and nonpoint source/stormwater sources. Connecticut has initiated a point source trading program for 79 municipal sewage treatment plants (STPs) to facilitate implementation of the TMDL wasteload allocation (WLA) and is investigating the potential for incorporating nonpoint source/stormwater into the existing Nitrogen Credit Exchange (NCE).

Several prerequisite conditions essential to the success of the current point source trading program have been met. Briefly, (1) all the STPs contribute to the same water quality problem; (2) the technology to remove N and meet the targets exists; (3) there are compelling member benefits to participate, especially cost savings; (4) sources can easily be monitored and tracked by end-of-pipe monitoring; (5) credit cost calculations are based on established and agreed upon protocols founded in state legislation; (6) sources of N are diverse and create viable supply and demand conditions while reducing overall cost, with close control by a Nitrogen Credit Advisory Board (NCAB); and (7) transaction costs are low relative to credit prices. In operation since 2002, the NCE has proven to be a viable and effective mechanism for meeting the nitrogen WLA.

The economic record of the NCE demonstrates the vigor of trading over the first five years of completed trades from 2002 to 2006 (Table 16). In sum, more than 10 million credits have been traded on the NCE, representing more than $22 million in economic activity.

The use of geographically-based trading ratios is instrumental to the relative cost of meeting N reduction limits at the 79 treatment plants, which are scattered throughout the state (Figure 21). Because N is reactive as it travels down rivers into the Sound, and the Sound's currents further affect relative impacts as they transport N and the resulting algal blooms to the hypoxic areas at varying efficiencies, location of each treatment plant makes a difference in relative impact on dissolved oxygen per pound of N discharged at end of pipe. Generally, the closer a POTW is to the edge of the Sound, and the closer to the hypoxic zone, the higher the trading ratio (Figure 22). For plants with high trading ratios, economics often favor treatment, while those with lower ratios may find the purchase of credits economically advantageous over treatment.

Table 16: Performance of the Nitrogen Credit Exchange

Trading Year	Credit Price (Dollars)	Purchased (Dollars)	Sold (Dollars)	Purchased (1000 Credits)	Sold (1000 Credits)
2002	$1.65	$1,317,223	$2,357,323	798	1,429
2003	$2.14	$2,116,875	$2,428,636	989	1,135
2004	$1.90	$1,786,736	$2,659,804	940	1,400
2005	$2.11	$2,467,757	$1,315,392	1,170	623
2006	$3.40	$3,828,114	$2,394,956	1,126	704
Total		$11,516,705	$11,156,111	5,023	5,291

Source: Connecticut Department of Environmental Protection, 2007

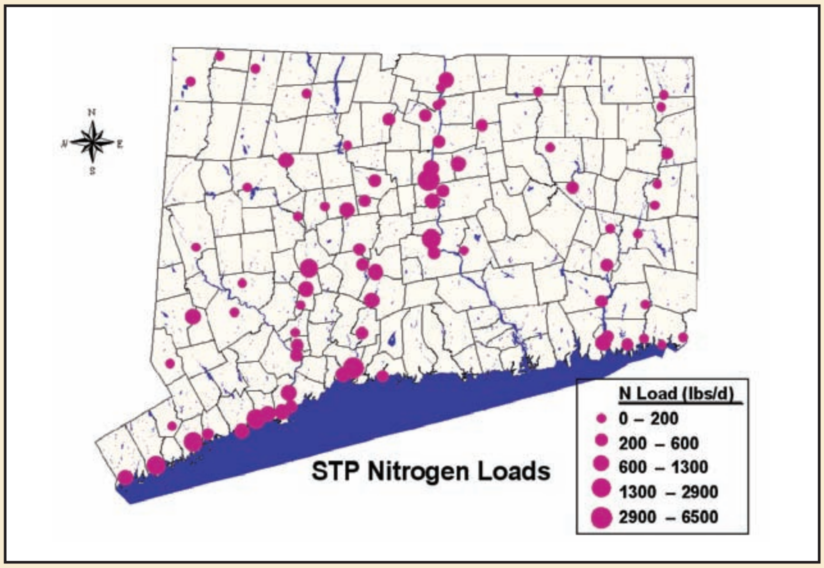

Figure 21: Relative nitrogen discharge (lbs/day) from 79 POTWs.

Source: Connecticut Department of Environmental Protection, 2007

The point source NCE does not reflect a free market approach to trading. Demand is set by the annual general permit limit and supply of credits is constrained by the availability of Clean Water Fund dollars and the timing and location of N removal projects. Credits are bought and sold from the state, thus the number of credits purchased does not need to match the number of credits sold (as would typically be true in a tradable permit system). Nevertheless, there is a tendency towards implementing cost effective projects as sewage treatment plant authorities decide whether it is less expensive to treat or buy credits, and try to predict when that break-even point might occur that would warrant application for project funding.

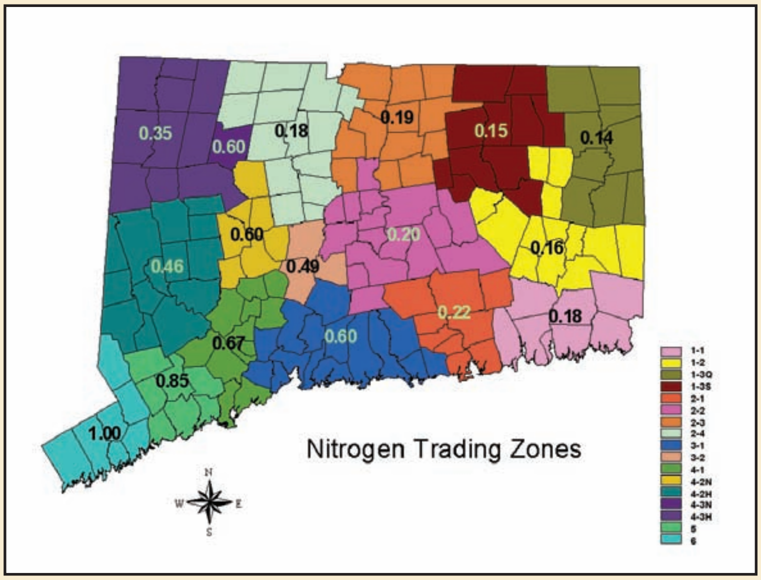

Figure 22: Trading ratios for municipalities in Connecticut.

Source: Connecticut Department of Environmental Protection, 2007

Incorporating a nonpoint source/stormwater (NPS/SW) component into the existing point source trading program presents some difficult challenges. Among the seven prerequisite conditions listed above that are well met by the current point source program, NPS/SW trading does not provide compelling economic benefits for members; NPS/SW N is difficult to quantify and track; credit cost estimation does not have a strong foundation in any existing programs; NPS/SW credit costs, though geographically diverse, may not result in significant implementation savings; and transaction costs (or time spent negotiating the ground rules for NPS/SW trading) may be considerably higher than for point source credits. Many of these obstacles can be overcome by deferring to models and textbook costs and efficiencies for NPS/SW BMPs. Tracking will still be a challenge because of the sheer number and distribution of BMPs that can be applied throughout the state.

Basic economic principles suggest that a free-market arrangement will not produce many NPS/SW credits for market. Costs are much higher than for point source credits and a regulatory approach must therefore be instituted to formalize the load allocation for nitrogen and to structure participation by municipalities.

If a NPS/SW trading component were to be added in the future, it would most likely also be an incentive-based program rather than a free-market approach. Nitrogen is difficult and costly to control in Connecticut's urban/suburban setting, and reductions are unlikely to be cost- competitive with POTW credits in a free market system. However, because municipalities are required to implement the Phase II stormwater permit, and various federal, state, and local programs require or emphasize NPS/SW management, there may be benefits for an incentive-based approach to offset some of those costs. For example, payment for NPS/SW reductions at the same credit prices paid to POTWs under the NCE would help defray costs and encourage additional nitrogen reductions from stormwater/NPS sources. Connecticut and the NCAB will continue to evaluate and explore the viability of these options.

Market approaches and trading can lower costs and increase economic efficiency of Nr control. The approaches may well have to be situation specific and depend on a structured regulatory framework to create the market or trading opportunity. As with the 1990 Amendments to the Clean Air Act, the design of market based instruments is a product of technical capability, regulatory design, and public preference. Implementation can be tedious but the benefits in efficiency substantial, even after being balanced against equity concerns. However, there can be something of a geographical and supply/demand mismatch between nonpoint sources and point sources that might be trading partners. Ribaudo and Nickerson identify only 142 of 710 eight-digit Hydrologic Watershed Units containing waters impaired by nitrogen where trading would be most likely (Ribaudo and Nickerson, 2009).

Further, the cost for management will be enormous. EPA's Clean Water Needs Survey (EPA, 2008b) has identified more than $200 billion in wastewater management infrastructure needs, and those needs do not fully address nutrient control from both traditional point and nonpoint/stormwater sources or consider alternative technologies.

Biophysical controls in terrestrial environmental systems

As indicated in Figure 2, approximately 36 Tg of new Nr is introduced into the U.S. each year. This new Nr is derived from sources that include consumption of ~11 Tg of synthetic N fertilizer, ~8 Tg of N that is fixed biologically by crops, and ~ 5 Tg that is emitted from fossil fuel combustion annually. This N is used to produce food and fiber (~15 Tg) or is formed during electrical generation, industrial production, or transportation. Efforts to decrease the creation of new Nr should first look to conservation.

Reduction in use of fossil fuel and/or decreased Nr emission can come through a variety of mechanisms such as more energy-efficient industrial processes, homes, and vehicles. Further gains are possible through conservation practices and alternatives to wasteful approaches, such as improving public transportation to minimize use of personal automobiles, and use of local products that do not require long-distance shipping.

Improvements in food and fiber production and changes in diet can also play an important role in limiting Nr. Because agriculture is the largest consumer and producer of Nr, consumption of fertilizer N could be decreased by changes in diet and increasing fertilizer N use efficiency in crop and fiber production systems. The control points discussed in this section include: protein consumption in the human diet; removing croplands that are highly susceptible to Nr loss from crop production; decreasing fertilizer N demand by increasing fertilizer use efficiency in crop and fiber production, as well as on residential and recreational turf grass; and better management of Nr in manure from livestock production in CAFOS.

Decreasing the amount of fertilizer N needed through changes in human diet

Along with increasing fertilizer N use, continued high intake of protein in developed countries and changes in the diet of people in developing countries will likely lead to greater N losses from global food production in the future. The first aspect of changes in food production concerns the increasing protein consumption that is occurring as global population increases and gets wealthier. This is likely to require increased N input into food production (Naylor et al., 2005; Galloway et al., 2007).

The average protein supply per person in developed countries is presently ~100 g per day, while in the developing countries it is only ~65 g per day (Food and Agricultural Organization Statistical Database (FAO FAOSTAT, 2010a). There is a direct proportionality between protein and nitrogen composition of food (ca 0.16 g N per 1 g protein). On average in 1995, developed countries consumed ~55% of total protein from animal sources while developing countries derived ~25% of total protein from animals. Protein consumption was highest in the U.S. and Western Europe, ~70 and ~60 g animal protein per person per day, respectively. In 2003, total protein consumption in the U.S. was 115 g per person per day (74 derived from animals and 41 from vegetable) (FAO FAOSTAT, 2010a). In developing countries, the greatest change in animal protein consumption has occurred in China where the consumption of meat products has increased 3.2 fold (from ~10 to ~32 g per person per day) since 1980. In Sub-Saharan Africa there has been no increase in either total (~50 g per person per day) or animal protein (~10 g per person per day) consumption during the past 30+ years (Mosier et al., 2002).

The reason for focusing on the consumption of animal protein is that more N is needed to produce a unit of animal protein than an equal amount of grain protein. Bleken et al. (2005) note that the N cost of animal production in Norway and the Netherlands was approximately five units of N in feeds for each unit of N produced. Approximately 2.5 units of N are required to produce a unit of wheat protein-N. Bequette et al. (2003) report that dairy cattle consume four units of N in feeds (including forage and grains) for every unit of N that appears in milk. Using a range of efficiencies for animal production practices, Kohn et al. (1997) estimated that 4 to 11 units of fertilizer N would be used in a whole farm system to produce a unit of milk protein. This ratio would be lower when using legume N to feed cattle, as is commonly done. Based upon the extra N required to produce animal protein compared to grains, continued high protein consumption in developed countries and changes to higher protein diets in developing countries will likely increase N input and losses in food production.

Moderating this increase by decreasing the average amount of total protein consumed in developed countries is one mechanism of limiting part of the expected increased N requirement in food production. One example of a country with a healthy diet and moderate consumption of animal protein is Italy in 1963. At that time, food supply was adequate to ensure sufficient nutrition to all groups of society (Bleken, 1997). Total protein consumption was 85 g per person per day, and consumption of animal protein was 32 g, roughly half of the current U.S. diet, and yet much higher than the average of developing countries. Another example is Japan, where animal protein consumption has traditionally been low, although it has increased from 25 g in 1963 to 54 g animal protein per person per day in 1995. In the same period the total protein consumption has increased from 73 g to 96 per person per day.

Bleken (1997) analyzed the relation between human diet and global N need for food production. Her analysis indicates that the total N needed for diets with high animal protein intake (comparable to many industrialized countries today) is almost twice as high as the N needed for the average diet in Italy 1963, or for Turkey in 1993. Based on her analysis, the Committee assumes that in the high-N input regions, per capita N need for food production may be reduced by 45%, which would reduce present-day N inputs by 15% worldwide.

Switching to a lower protein diet may not, however, reduce N losses if the new diet includes increased quantities of fruits, vegetables, and nuts, in addition to staple grains, beans and pulses. Vegetables, fruit, and nuts are high value crops that typically require large inputs of fertilizers and pesticides when produced at a large, commercial scale, and N fertilizer losses can be considerably larger than for grain crops. Having a very diverse diet that includes a wide range of high-value fruits and vegetables available year round (whether they are in-season locally or not) also has consequences for N inputs/outputs from agriculture – both within the U.S. and globally. EPA and USDA are encouraged to develop programs that stress how both human health and environmental health will improve with a greater focus on the human diet. It has been estimated that 30% – 40% of the food prepared for consumption in the U.S. is wasted (Kantor et al., 1997; Hall et al., 2009). Thus, additional Nr may be conserved by decreasing the amount of food that is wasted.

Removing croplands that are susceptible to Nr loss from crop production

An analysis of NO_3^- loading in the Mississippi River Basin (Booth and Campbell, 2007) provides estimates of N input from agricultural lands. Similar estimates were provided by Del Grosso et al. (2006). Recommendations in this analysis are essentially the same as those arrived at in the original national hypoxia assessment, which suggested that the most leaky lands be taken out of production (Doering et al. 1999). Booth and Campbell state:

> *Nitrogen derived from fertilizer runoff in the Mississippi River Basin (MRB) is acknowledged as a primary cause of hypoxia in the Gulf of Mexico. To identify the location and magnitude of nitrate runoff hotspots, and thus determine where increased conservation efforts may best improve water quality, we modeled the relationship between nitrogen inputs and spring nitrate loading in watersheds of the MRB. Fertilizer runoff was found to account for 59% of loading, atmospheric nitrate deposition for 17%, animal waste for 13%, and municipal waste for 11%. A nonlinear relationship between nitrate flux and fertilizer N inputs leads the model to identify a small but intensively cropped portion of the MRB as responsible for most agricultural nitrate runoff. Watersheds of the MRB with the highest rates of fertilizer runoff had the lowest amount of land enrolled in federal conservation programs. Our analysis suggests that scaling conservation effort in proportion to fertilizer use intensity could reduce agricultural nitrogen inputs to the Gulf of Mexico, and that the cost of doing so would be well within historic levels of federal funding for agriculture. Under this simple scenario, land enrolled in conservation programs would be increased by about 2.71 million hectares, a 29% increase over 2003 enrollments, while land taken out of traditional fertilized agriculture and enrolled in conservation programs would constitute about 3% of 2003 fertilized hectares.*

The Booth and Campbell approach places the leakiest intensively cropped lands into government programs like the Conservation Reserve Program – where they would be put into grass or cover crops. Doering et al. (1999) had a somewhat different approach. Under their analysis, nitrogen use or nitrogen loss reductions were imposed on agriculture, and the U.S. Agricultural Sector Mathematical Programming (USMP) model adjusted crop rotations, tillage practices and fertilizer inputs within the Mississippi Basin – meeting the given Nr constraint while maximizing producer and consumer welfare. The model favored those crops and cropping systems at different points in the landscape having low nitrogen leakage. Where the model could not find a crop production system having positive returns while meeting the Nr restrictions, the land was retired from production. This analysis suggests opportunities for maintaining land in agricultural production while still reducing Nr losses through better matching of land characteristics with crops and cropping systems.

This 1999 analysis of the Mississippi Basin was carried out in the context of cost effective approaches – starting with the most cost-effective (in terms of producer and consumer welfare) and moving to less cost-effective approaches as more and more nutrients were controlled. This included both restriction of fertilizer inputs, buffers, and wetland remediation as well as the land use changes and crop rotations referred to above. The suggestions presented by the Committee for Nr reductions that could be achieved from agriculture with existing technology are consistent with the cost effective approaches in the 1999 Hypoxia Assessment's economic analysis. Cost effectiveness and alternative cropping systems were considered in the SAB report, *Hypoxia in the Northern Gulf of Mexico: An Update by the EPA Science Advisory Board* (U.S. EPA SAB, 2007) but unfortunately as pieces from individual study examples rather than as an integrated approach like the 1999 Hypoxia Assessment (Doering et al., 1999).

Decreasing fertilizer N demand by increasing fertilizer use efficiency in crop and fiber production

The largest input of Nr in North America is N fertilizer used for crop production. The mean annual N fertilizer input to North America between 1999 and 2003 was 12.5 Tg. Of this fertilizer N, 66% was used to fertilize cereal crops, mainly corn and wheat (Dobermann and Cassman, 2005).

As previously discussed, corn yield in the U.S. has increased (from an average of 100 bushels per acre in 1985 to 136 bushels per acre in 2005) as a result of improved nutrient and pest management, expansion of irrigated area, conservation tillage, soil testing, and improved crop genetics (yield and pest resistance) (Council for Agricultural Science and Technology [CAST], 2006). From 1980 to 2000, N-fertilizer use efficiency (NFUE, kg grain produced per kg applied N, or kg grain / kg N) increased from 42 to 57 kg grain / kg N, a 35% efficiency gain during a period when average U.S. corn yields increased by 40% (Fixen and West, 2002). Despite this steady increase in NFUE, the average N fertilizer uptake efficiency for corn in the north-central U.S. was 37% of applied N in 2000 based on direct field measurements (Cassman et al. 2002). These results indicate that greater than 50% of applied N fertilizer is vulnerable to loss pathways such as volatilization, denitrification, runoff, and leaching. The results also suggest there is substantial room for improvement in N efficiency currently achieved by farmers. Although progress has been made to increase both cereal yield and NFUE, a concerted effort to further increase NFUE remains a logical control point to reduce production costs, because N fertilizer represents a significant input cost, and to limit Nr leakage (e.g., NH_3, NO_x, N_2O, NO_3^-) from agroecosystems.

The goal of reducing Nr while sustaining adequate rates of gain in cereal production to meet expected food demand will require increases in NFUE, which in turn will require innovative crop and soil management practices. This need is exacerbated by the recent increase in demand for corn to produce ethanol biofuel. The concept of improved N synchrony (practices that better match the amount, timing, and geospatial location of applied N to crop-N demand and the N supply from indigenous soil resources) is generally viewed as the most appropriate approach for improving NFUE (e.g., Appel, 1994; Cassman et al., 2002). The challenge is to attain greater synchrony between crop N demand and the N supply from *all sources* (e.g., soil, fertilizer, organic inputs such as manure, compost, or green manures) throughout the growing season. Losses from all N-loss mechanisms increase in proportion to the amount of available N present in the soil profile at any given time.

Several promising technologies and combinations of technologies have emerged in recent years. Significant increases in NFUE are often achieved through reducing N fertilizer use by 10 to 30%, while still maintaining or even slightly increasing yields (Giller et al. 2004). Figure 23 indicates where the greatest gains in NFUE are expected to be realized from investments in different technology options. Improvements in crop and soil management practices will contribute to higher NFUE by achieving greater congruence in timing of the supply of applied N with crop-N demand and the N supply from indigenous soil resources. While there is relatively small scope for specific biotechnology traits to improve NFUE, overall improvement in crop genetics from commercial breeding efforts that focus on increasing yield and yield stability will continue to play a significant role in improving overall NFUE. However, large investments in research, extension education, and technology transfer will be required, and significant incentives must be implemented, to achieve the degree of improved synchrony needed to make substantial improvements in NFUE. The need to accelerate the rate of gain in crop yields to meet increasing demand for human food, livestock feed, and biofuels represents an additional new challenge. Crop prices are expected to rise as they more closely track the price of petroleum (Council for Agricultural Science and Technology, 2006). Higher crop prices will motivate farmers to achieve higher yields, and higher crop yields require a greater amount of N uptake to support increased biomass production (Greenwood et al., 1990). Therefore, an explicit emphasis on developing technologies that contribute to both increasing yields and NFUE will be needed to ensure that the goals of food security, biofuel production, and protection of environmental quality are met.

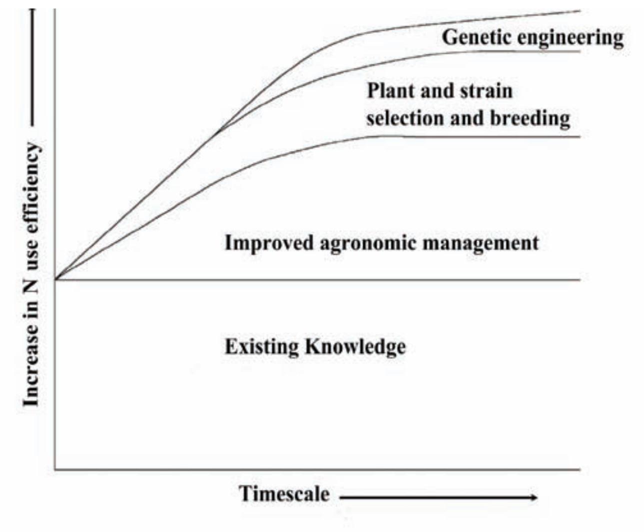

Figure 23: The likely impact of research investment in increasing N fertilizer use efficiency

Source: Giller et al., 2004 (Figure 3.2, p. 48). Reprinted with permission from Island Press; copyright 2004, SCOPE.

Alternatives to current urban landscaping practices

Section 2.2.4 discussed the use of turf grasses as a prominent feature in U.S. urban landscapes with over 1 TgN used to fertilize lawns each year (Table 9). New developments are most amenable to landscaping practices that may minimize the need to use supplemental fertilizer. These practices include preservation of the natural soil profile, use of turf types that require little or no fertilizer, minimizing turf areas, using organic maintenance techniques, and choosing alternatives to lawns and exotic plant species such as naturalistic landscaping. Many of these practices are part of a low impact development philosophy, which can also combine other best management practices to mitigate the effects of impervious cover and landscape changes. Existing development is also amenable to many of these practices, especially conversion of typical residential and commercial lawns to natural landscapes and retrofitting other BMPs that promote infiltration, such as rain gardens.

Structural and non-structural Best Management Practices (BMP) to treat runoff

There are probably hundreds, if not thousands, of BMPs that have been designed and manufactured to treat runoff from both urban and agricultural lands. Whether applied to new development or existing agricultural or urban land use, most follow basic principles that simulate natural land features and processes that remove pollutants from runoff. They promote infiltration to take advantage of the cleansing value of passage through soils and to reduce runoff volumes, and provide for biological or chemical conditions that help remove pollutants (NRC, 2008b, 2009).

The most notable of the processes for managing Nr is providing conditions that are adequate to denitrify Nr in the waste-stream in a process called biological nitrogen removal (BNR). BNR simply creates conditions that convert initial forms of nitrogen to nitrate via oxidation, and convert nitrate to dinitrogen gas by providing conditions (especially high carbon and low oxygen) where the denitrification process can occur. These simulate natural conditions such as nitrification that occurs in oxic soils as water-borne nitrogen infiltrates into the soils and groundwater, and denitrification that occurs in highly-organic, saturated soils such as in wetlands where oxygen is low.

Most BMPs are considered structural, and may be highly engineered "package" plants that can treat sewage or runoff, depending on scale and structure, or simple detention basins that allow sediments and adhered pollutants to settle out. "Artificial" wetlands are a good example of a more sophisticated BMP that takes advantage of natural processes, and may be created at the end of the stormwater pipe, or at edge of field. Structural BMPs are an important part of any strategy to limit reactive nitrogen loss to the environment. For example, The State of Iowa contains some of the most productive agricultural land in the world. Of the 36 million acres of land, 23 million acres are planted in corn or soybeans. Approximately 39 percent of the corn/soybean acres are drained with an estimated 800,000 miles of tile (Cutler, 2000). Each year, thousands of miles of new or replacement tile are installed. This drainage network is responsible for the conveyance of 90 percent (Crampton et al., 2006) of the nitrate that appears in Iowa's surface waters. Control of nitrogen discharge from drainage tile will be needed to limit reactive nitrogen loss to the environment from agriculture.

Various approaches have been proposed with varying degrees of success. Constructed wetland, bio-reactors, and drainage tile encapsulated with biomass have proven to reduce 90 percent of the reactive nitrogen loss to the environment (Blowes et al., 1994). It would seem reasonable to require any new or replacement drainage tile to implement a control strategy at the time of construction and a retrofit program for the remaining drainage systems. With an average of two years to pay back the cost of drainage systems due to increased crop yield, the added cost for nitrogen control does not seem to be unreasonable.

Non-structural BMPs are often preservation actions, as discussed earlier, or activities that prevent pollutants from entering the waste stream such as street sweeping or fertilizer limitation.

Engineered and restored wetlands to decrease NO_3^- loading of aquatic systems

The construction and/or restoration of wetlands have received considerable attention in the past two decades as a conservation method. Such an approach has several positive attributes including promoting denitrification in watersheds containing or receiving Nr, flood protection, habitat preservation, and recreational potential (Hey and Philippi, 1995). In the upper Mississippi basin optimum siting of wetlands could result in as much as 0.4Tg of NO_3^- converted to N_2 (Mitsch et al., 1999, 2001; Hey, 2002).

Much of the nitrate leached from agricultural fields could be removed from drainage water in natural, created, or restored wetlands. Nitrate removal from the water column in wetlands is performed by plant uptake, sequestration in the soils, and microbial transformation that includes immobilization and denitrification. Plant uptake and microbiological immobilization result in temporary storages in the system since most nitrogen will eventually return to the wetland via plant death and decomposition. In contrast, denitrification can constitute a real nitrogen sink because NO_3^- is converted mainly to N_2 that is emitted to the atmosphere (Clement et al., 2002). As discussed in Section 4.7, the potential for the formation of N_2O is of concern if such systems are not operated properly.

In addition to preserving existing wetlands, there are two other basic approaches that utilize wetlands to reduce the Nr and other nutrients reaching rivers, streams, and vulnerable downstream coastal systems. These approaches are: 1) creation and restoration of ecosystems, principally wetlands and riparian forests, between farms and adjacent ditches, streams and rivers; and 2) diversion of rivers into adjacent constructed and restored wetlands all along the river courses (Mitsch and Jorgensen, 2004; Mitsch and Gosselink, 2007).

The Committee notes that if wetlands can be economically and effectively restored where croplands now exist on hydric soils within the 100-year floodplain, this may be an important NO_3^- control mechanism. Cropland on hydric soil in the floodplain occupy about 6.9 million acres (2.8 million hectares). If this area and its wetlands were given back to the Mississippi River, over a million tons of NO_3^- -N would be annually removed or prevented from reaching the Gulf of Mexico (Mitsch et al., 1999; Hey, 2002; Hey et al., 2004). To give scale to the solution needed, restoration of over 4.9 million acres (2 million hectares) of wetlands is needed in the Mississippi-Ohio-Missouri (MOM) river basin to reduce the nitrogen load to the Gulf of Mexico sufficiently to ensure a reduction in the size of the hypoxic zone in the Gulf of Mexico (Mitsch et al., 2001).

At a series of workshops on restoration of the MOM river basin in 2003-2004 (Day et al., 2005; Mitsch and Day, 2006), scientists and managers were asked to focus on needed research and chokepoint opportunities for managing N in that basin. They concluded that a major, interdisciplinary research program (as a lead-in to the actual restoration of wetlands and rivers) was needed, with sufficient funding, study sites, and time to reduce remaining uncertainties about the efficacy of wetlands to solve pollution problems related to N. It was recommended that, to implement this program, 20 to 30 full-scale, existing and new agricultural/wetland demonstration projects should be located throughout the country and instrumented to study agricultural runoff into wetlands in a variety of soil conditions. Pilot and full-scale studies of diversions into riparian systems along river channels were recommended in order to determine the effectiveness of these systems for nutrient removal. The Committee notes that these research and demonstration projects have not been undertaken, and that there is a continuing need for this work.

Further illustration of the use of wetlands as a tool for Nr management tool is presented in Appendix C, Water Quality Trading in the Illinois River Basin (D. Hey, personal communication).

Technical controls (control points) on transfer and transformation of atmospheric emissions of Nr in and between environmental systems: NH_3

Newly fixed Nr is produced biologically or added as fertilizer to meet the demand for food and fiber production. Much of the N is used in cereal crop production and cereal crops are then used to feed livestock. The new Nr is then recycled through the livestock production system where it becomes again susceptible to losses to the atmosphere as ammonia and NO_x, and is available for additional N_2O production and movement into aquatic systems as NH_4^+ and NO_3^-.

The bulk of the N fed to livestock ends up in manure, and where this manure (approximately one-half in urine and one-half in feces) is produced, there is often a much greater supply than can be efficiently or economically used as fertilizer on crops. For large concentrated animal feeding operations there is considerable expense associated with disposal of the manure. Various storage systems have been developed to deal with this excess manure, the most interesting of which, from the standpoint of integrated policy on N, convert the urea to N_2. The fraction of manure N that can be and is converted to N_2 remains a major unanswered scientific or technical question.

The NRC (2003) noted the paucity of credible data on the effects of mitigation technology on rates and fates of air emissions from CAFOs. The report did, however, call for the immediate implementation of existing atmospheric emission technology. The NRC (2003) also called for a mass balance approach in which the losses of N species such as NH_3, NO, N_2, and N_2O are expressed as a fraction of the total N loss. Quoting from the NRC report:

> *Storage covers for slurry storage tanks, anaerobic lagoons, and earthen slurry pits are being studied as a method to decrease emissions from those containments. Both permeable and impermeable covers are being studied. Tested covers range from inexpensive material such as chopped straw (on slurry containments only) to more expensive materials such as high density polyethylene. Covers can decrease emissions from storage but their net effect on emissions from the system is conditional on how the effluent is used on the farm.*
>
> *Anaerobic digestion in closed containment has been studied for many types of applications. Anaerobic digestion is the process that occurs in an anaerobic lagoon. When conducted in closed vessels, gaseous emissions including methane, carbon dioxide and small amounts of other gasses (possibly ammonia, hydrogen sulfide, and VOCs) are captured and can be burned for electricity generation, water heating, or simply flared. The in-ground digester being tested on a swine farm in North Carolina is an example of the ambient temperature version of this technology (there are also mesophilic and thermophilic designs). The concentration of ammonia remaining in effluent from that digester is higher than the concentration in lagoon effluent and can be volatilized once exposed to air.*

Recent research (e.g., Bicudo et al., 2004; Funk et al., 2004a,b; Shores et al., 2005) demonstrates reduction in NH_3 emissions after a permeable cover was installed. Miner et al. (2003) reported that a polyethylene cover can reduce NH_3 emissions by ~80%, but it is not clear what fraction of that N was converted to N_2. Harper et al. (2000) reported that in a well-managed swine lagoon denitrification N_2 losses can be equivalent to N lost as NH_3, in other words about 50% efficiency. Kermarrec et al. (1998) reported that sawdust litter helped reduce NH_3 emissions from pig manure with 44-74% of manure

N converted to N_2, but greater than 10% of the manure N was released as N_2O. Sommer (1997) found that NH_3 was emitted from cattle and pig slurry tanks at the rate of 3.3 kg N m^2yr^{-1} until covered with straw. After straw application NH_3 emissions were below detection limit. Mahimairaja et al. (1994) reported that NH_3 volatilization was reduced by 90-95% under anaerobic conditions. Section 2.2.4 contains a discussion of best management practices to minimize NH_3 emissions from livestock waste, and presents finding and recommendation 6 on the need for a framework for manure management.

Technical controls (control points) on transfer and transformations of atmospheric emissions of Nr in and between environmental systems: NO_x

As previously discussed, a major contributor to Nr in the atmosphere is fossil fuel combustion. During the combustion process NO_x ($NO_x = NO + NO_2$) are released to the atmosphere. Globally the production of NO_x has accelerated in the last few decades, primarily through the increase in fossil fuel combustion (Galloway et al., 1995; 2008). With this increase in emissions from ~5 Tg N in 1940 to ~ 25 Tg N in 2005, combustion of fossil fuels accounted for about 50% of the total global NO_x emissions for 1990. Of the anthropogenic sources, fossil fuel, aircraft, biomass burning, and part of the soil emission are most important (Holland et al., 1997). Although global NO_x emissions continue to increase, these emissions are declining in the U.S. (see Section 2.2.1).

Nitrogen oxide is formed during combustion by three mechanisms:

- Thermal NO_x where N_2 and O_2 combine at high temperatures (thermal pathway dominates at temperatures greater than approximately 1500 C) to form NO through the Zeldovich mechanism
- Fuel NO_x where nitrogen from a fuel (e.g., coal and biofuels) is released as some intermediate and then combines with O_2 to form NO
- Prompt NO_x where N_2 reacts with hydrocarbon radicals in flames, forming various compounds including hydrogen cyanide and other cyano radicals. These in turn form NO_x. Contributions of prompt NO_x are usually low as compared to fuel NO_x.

There are several ways to control emissions of NO_x. The most common controls are on coal-fired electric utility generators and those are discussed below. Following the discussion of electric utility generator controls, or external combustion systems, there is a discussion on internal combustion controls.

Reduction of the temperature limits the kinetics of the N/O_2 reaction. Temperature can be controlled by using a fuel-rich mixture versus fuel lean. In this case the reactions take place at lower temperatures. Fuel-rich mixtures also reduce the amount of O_2 available for reaction and there are changes in the chemical mechanisms which limit the oxidation of N_2. If fuel-lean mixtures are used for temperature control, while the temperature is lower there is a significant amount of O_2 present. Typically, in external combustion systems controls are implemented by using less excess air and using staged combustion. In addition, flue-gas recirculation (FGR) is used to lower the temperature. Low-NO_x burners operate under the principle of internally staging the combustion. To reduce fuel NO_x, air and fuel staging are used to reduce the peak temperature where air and fuel are admitted in separate locations.

Chemical reduction of NO_x is also possible. These methods include: selective non-catalytic reduction (SNCR), selective catalytic reduction (SCR), and fuel reburning. SNCR is an add-on technology where urea or NH_3 is injected in a controlled temperature zone to allow the reduction of NO_x. SCR is also an add-on technology where the flue gas must pass through a catalyst bed to allow reaction between ammonia and NO_x. Care must be taken with both technologies to avoid NH_3 slip. Fuel reburning requires the injection of a fuel to create a zone where NO_x is reduced to N_2. Low NO_x burners may also use an internal fuel reburning to reduce the NO_x.

For internal combustion engines, the same mechanisms discussed above are used in a variety of different ways, since these systems are using high pressure and predominately have thermal NO_x versus fuel NO_x formation. Most technologies involve the need to reduce the peak temperature and duration of high temperatures of the combustion zone. For example, gas turbines utilize low NO_x burners, while spark ignition engines utilize a three-way catalyst which requires less than 0.5% O_2. In this case, additional NO_x is reduced by utilizing unburned fuel as a reagent over the catalyst for chemical reduction of NO_x. It should be noted, however, that a side reaction for the three-way catalyst system produces ammonia. NO_x emissions can be reduced in diesel engines by delaying the injection of the fuel, and by retarding the timing in spark-ignited engines. Engines also use exhaust gas recirculation (EGR) to reduce the peak temperatures. Recent road side studies have indicated high efficiency (~90%) for NO_x removal from the American light-duty fleet (Bishop and Stedman, 2008).

6

SAB Findings and Recommendations for Nr Data Collection, Risk Management, and Research

This concluding chapter contains the Integrated Nitrogen Committee's findings and its recommendations to EPA. Section 6.1 discusses the need for a comprehensive program to monitor reactive nitrogen. Section 6.2 provides the Committee's overarching recommendations to EPA. Section 6.3 contains suggestions for near-term actions that might be taken by EPA and other management agencies to decrease Nr entering the environment from various sources. Section 6.4 contains specific findings and recommendations corresponding to each of the Committee's four study objectives.

The first objective of the study was to identify and analyze, from a scientific perspective, the problems Nr presents in the environment and the links among them. To accomplish this objective, the Committee examined the flows of Nr within the food, fiber, feed and bioenergy production systems and developed lands in the U.S., paying special attention to the locations within each of these systems where Nr is lost to the environment. The same process was employed for fossil fuel energy production but, since all the Nr formed and released during energy production is lost to the environment, the Committee identified the important energy producing sectors that contribute to Nr emissions. The Committee found that agriculture and domestic use of fertilizers to produce food, feed, and fiber (including bioenergy and BNF) and combustion of fossil fuels are the largest sources of Nr released into the environment in the U.S.

The Committee also examined the fate of the Nr lost to the environment, estimated the amount stored in different systems (e.g., forest soils) and tracked Nr as it is transferred from one environmental system (e.g., the atmosphere) to another (e.g., terrestrial and aquatic ecosystems). Source and fate analyses set the stage for identifying the environmental and human health problems Nr presents, and the links among them. Using the nitrogen cascade, the Committee identified the impacts Nr has on people and ecosystem functions as it moves through each system and contributes to adverse public health and environmental effects, including photochemical smog, nitrogen-containing trace gases and aerosols, decreased atmospheric visibility, acidification of terrestrial and aquatic ecosystems, eutrophication of coastal waters (i.e., harmful algal blooms, hypoxia), drinking water concerns, freshwater Nr imbalances, GHG emissions and subsequent climate change, and stratospheric ozone depletion.

The second objective of the study was to evaluate the contribution an integrated N management strategy could make to environmental protection. To accomplish this objective the Committee identified actions that could be taken to better manage Nr. These actions take into account the contributions of all Nr sources and chemical species that adversely impact human health and environmental systems, and the need to ensure that solving one problem related to Nr does not exacerbate another problem or diminish ecosystem services that support societal demands.

The third objective of the study was to identify additional risk management options for EPA's consideration.In addressing this objective, the Committee identified four major goals for management actions that collectively have the potential to decrease Nr losses to the environment by about 25%. Decreasing Nr emissions by these actions will result in further decreases in Nr-related impacts throughout the nitrogen cascade. The Committee has suggested a number of ways to attain these management goals, including conservation measures, additional regulatory steps, voluntary actions, application of modern technologies, and end-of-pipe approaches. The Committee notes that these are initial but significant actions; however, others should be taken once the recommended actions are completed and assessed, and further opportunities are explored in an adaptive management approach.

The fourth objective of the study was to make recommendations to EPA concerning improvements in Nr research to support risk reduction. The Committee has provided numerous recommendations for additional Nr research to support risk reduction activities.

6.1. Need for Comprehensive Monitoring of Nr

In previous sections of this report the Committee has discussed the importance of monitoring reactive nitrogen in the environment. The Committee recommends establishing a program for comprehensive monitoring of the multiple forms of reactive nitrogen as both stocks and flows as they pass through different media and ecosystems. There are two major reasons for this overarching recommendation. The first purpose of monitoring is to provide the observational data on trends that will inform research into the complexity of the nitrogen cascade to better identify the most effective intervention points to reduce damage to human health and the environment by reactive nitrogen. The second need for monitoring is to be able to assess the effectiveness of policy interventions over time, and to apply the principles of adaptive management. As it becomes clear which strategies and policy instruments are effectively reducing the amounts of Nr entering the environment, and which are ineffective, it will be necessary to modify those interventions in response to the monitoring data. As conditions change (e.g., shifts in the technology of electric power production, new fuels for transportation, changing land use patterns and climate change), the nitrogen cascade will be modified, and the relative importance of sector specific policies will change. Only through comprehensive monitoring will it be possible to manage Nr effectively.

Finding 20: The Committee has determined that an integrated approach to monitoring that includes multiple media (air, land, and water) components and considers a suite of environmental and human concerns related to reactive nitrogen in the environment (e.g., Nr effects, climate change, human health) is needed. Some of the phenomena presented in this report need more definition and verification but, more importantly, as controls are brought to bear on Nr, improvements need to be measured to verify and validate successful management strategies. If the desired improvements are not realized as shown by the collected data, corrective measures will be required. Such an adaptive approach acknowledges the likelihood that management programs will be altered as scientific and management understanding improve.

Recommendation 20: *The Committee recommends that EPA initiate discussions and take action to develop a national, multi-media monitoring program that monitors sources, transport and transition, effects using indicators where possible, and sinks of Nr in keeping with the nitrogen cascade concept. This comprehensive program should build upon existing EPA and state initiatives as well as monitoring networks already underway in other federal agencies such as the U.S. Geological Survey programs and the NADP effort.*

6.2. Overarching Recommendations

Human activities have significantly increased the introduction of Nr into the U.S. environment and, through radical alterations of land use, have eliminated many of the natural features that once may have provided pollutant treatment. While there have been significant benefits resulting from food production, there have also been, and continue to be, major risks to the health of both ecosystems and people due to the introduction of Nr into the nitrogen cascade.

In its 1990 report, *Reducing Risk,* the Science Advisory Board recommended that the EPA increase its efforts to integrate environmental considerations into broader aspects of public policy in as fundamental a manner as are economic concerns. Other federal agencies often affect the quality of the environment, e.g., through the implementation of tax, energy, agricultural, and international policy, and EPA should work to ensure that environmental considerations are integrated, where appropriate, into the policy deliberations of such agencies. In the current era of increasing responsibilities without commensurate budgets, intergovernmental cooperation, partnerships and voluntary programs have become vital tools for agencies needing to stretch their resources to fulfill their missions.

Optimizing the benefits of Nr, and minimizing its impacts, will require an integrated nitrogen management strategy that not only involves EPA, but also coordination with other federal agencies, the States, the private sector, universities, and a strong public outreach program. The Committee understands that there are real economic costs to the recommendations contained in this report. For each recommendation there will of necessity be tradeoffs derived from the varying cost-effectiveness of different strategies.

The Committee makes four overarching recommendations:

Overarching Recommendation 1

The Committee recommends an integrated approach to the management of Nr. This approach will likely use a combination of implementation mechanisms. Each mechanism must be appropriate to the nature of the problem at hand, be supported by critical research on decreasing the risks of excess Nr, and reflect an integrated policy that recognizes the complexities and tradeoffs associated with the nitrogen cascade. Management efforts at one point in the cascade may be more efficient and cost effective than control or intervention at another point. This is why understanding the nature and dynamics of the N cascade is critically important.

Overarching Recommendation 2

The framing of the reactive nitrogen cascade provides a means for tracking nitrogen as it changes form and passes through multiple ecosystems and media. This complexity requires the use of innovative management systems and regulatory structures to address the environmental and human health implications of the massive amounts of damaging forms of Nr. It is difficult to create fully effective regulations de novo for such a complex system so we recommend utilizing adaptive management to continuously improve the effectiveness and lower the cost of implementation policies. This in turn will require a monitoring system that will provide feedback on the effectiveness of specific actions taken to lower fluxes and concentrations of Nr.

Overarching Recommendation 3

EPA should form an intra-Agency Nr management task force that will build on existing Nr research and *management capabilities within the Agency. This intra-Agency task force should be aimed at increasing scientific understanding of: (1) Nr impacts on terrestrial and aquatic ecosystems, human health, and climate,(2) Nr-relevant monitoring requirements, and (3) the most efficient and cost-effective means by which to decrease various adverse impacts of Nr loads as they cascade through the environment.*

Overarching Recommendation 4

Successful Nr management will require changes in the way EPA interacts with other agencies. To coordinate federal programs that address Nr concerns and help ensure clear responsibilities for monitoring, modeling, researching and managing Nr in the environment, the Committee recommends that EPA convene an Inter-agency Nr management task force. It is recommended that the members of this inter-agency task force include at least the following federal agencies: U.S. Department of Agriculture, U.S. Department of Energy, U.S. Department of Housing and Urban Development, U.S. Department of Transportation, National Oceanic and Atmospheric Administration, U.S. Geological Survey, U.S. Forest Service, and Federal Emergency Management Agency . This task force should coordinate federal programs that address Nr concerns and help ensure clear responsibilities for monitoring, modeling, researching, and managing Nr in the environment. The EPA Office of International and Tribal Affairs should work closely with the Department of State to ensure that EPA is aware of international efforts to control Nr and is developing national strategies that are compatible with international initiatives.

Similar recommendations for coordination and joint action among and between agencies at both state and federal levels have been made in the National Research Council's recent reports on the Mississippi Basin (NRC, 2008b, 2009). These intra and inter-agency Nr-management task forces should take a systems approach to research, monitoring, and evaluation to inform public policy related to Nr management. The Committee proposes that the intra and inter-agency task forces coordinate the following activities to implement a systems approach to Nr management.

Development of methods

Implementation of a systems approach will require development of methods to facilitate various aspects of Nr management. These include methods for: (1) establishing and evaluating proposed Nr budgets; (2) using life cycle accounting approaches for Nr management; (3) gathering and using data on N fertilizer use and other Nr sources and fluxes as the basis for informed policies, regulations and incentive frameworks for addressing excess Nr loads; (4) evaluating the critical loads approach to air and water quality management; (5) identifying and using indicators of excess Nr's economic damage and effects on human health and the environment; and (6) using systems-based approaches for controlling Nr releases to the environment.

Implementing best management practices (BMPs)

It will be necessary to improve the scientific understanding of BMPs that can be used for specific applications to manage Nr. In particular, this includes better scientific understanding of: (1) Nr requirements in agriculture to ensure adequate food, feed, fiber, and bioenergy feedstock supply while also avoiding negative impacts on the environment and human health; (2) Nr requirements for urban landscapes (e.g., residential and commercial) and their maintenance while avoiding negative impacts on the environment and human health; (3) planning and pollution prevention, including low impact development and natural ecosystem service preservation; (4) use of natural land features and attributes, such as wetland preservation and enhancement, natural soil profiles, and buffer strips; and (5) improved removal of Nr from sewage waste steams at both large-scale wastewater treatment facilities and individual subsurface (septic) systems. In addition, proactive extension and technology transfer approaches will need to be established to facilitate adoption of BMPs.

Developing appropriate tools and metrics for assessing impact from adoption of best management practices

Assessment activities will also be an important element of the systems approach to managing Nr. These activities should include: (1) quantifying the effectiveness and impact of policies and regulations focused on reduction of negative environmental impacts from Nr; (2) assessing combined carbon (C) and Nr effects on terrestrial and aquatic ecosystems; (3) assessing indicators/endpoints, costs, benefits, and risks associated with impairment of human health and decline and restoration of ecosystem services; (4) reviewing existing and proposed legislation for purposes of better integrating or designing regulatory activities that recognize the nitrogen cascade or streamlining procedures for enacting Nr risk reduction strategies; and (5) evaluating economic incentives, particularly those that integrate air, aquatic, and land sources of excess Nr.

Education, outreach, and communication

It will be necessary to develop new education, outreach, and communication initiatives. As discussed in this report, this includes a range of targeted outreach and education programs to manage Nr and achieve desired environmental outcomes.

6.3. Near-term Management Goals

The Committee puts forward four goals for actions that could be taken by EPA or other management agencies to decrease Nr entering the environment from various sources. We believe these goals can be attained over the near term (approximately 10-20 years) using existing and emerging technologies and practices. These suggestions, if implemented, have the potential to reduce total Nr loadings to the environment in the U.S. by approximately 25% below current levels. The Committee believes that these represent realistic and attainable near-term goals, however further reductions are undoubtedly needed for many N-sensitive ecosystems and to ensure that health-related standards are maintained. The Committee understands that actual policy decisions on the implementation of programs to limit Nr releases to the environment may differ from those listed below for a variety of reasons, but believes that an aggressive level of action, as represented by these goals, is critical given the growing demand for food and fiber production and energy use from population pressures and economic growth. The rationale for these goals is set forth below, along with recommended management options for achieving the goals.

Management Goal 1. Controls on NO_x emissions from mobile and stationary sources

The Clean Air Act (1970) and its Amendments (1990) have resulted in NO_x emissions that are less than 50 percent of what they would have been without existing controls. While this is an admirable accomplishment, there is still a need to seek improvements. NO_x emissions are an order of magnitude greater than at the beginning of the twentieth century. As a consequence, there remain significant negative impacts on both humans and ecosystems. In 2002, coal-fired utilities generated approximately 1.3 Tg N annually (see Figure 3). If all coal-fired plants used state-of-the-art NO_x controls, this number could be reduced by 0.6 Tg N/yr (calculations performed by Cohen, 2008); in fact, 2008 emissions have been reduced by 0.3 Tg N/yr from 2002 levels (see Figure 3), so in essence, half the reduction has already been accomplished. The EPA should continue to reduce NO_x emissions from major point sources, including electric generating stations and industrial sources, expanding the use of market mechanisms such as cap and trade. Under this scenario, it is likely that high-efficiency, low-emission power plants will be built for energy needs. Some controls on NO_x emissions are implemented only in the ozone season (May to September) (U.S. EPA 2009c). To protect welfare and avoid adverse effects on ecosystems, NO_x emissions controls should be implemented year round.

For mobile sources, emissions for highway and off-highway sources are approximately 2.2 Tg N/yr and 1.2 Tg N/yr, respectively. EPA is in the process of implementing a number of regulations that will reduce NO_x from mobile sources (see Appendix F). The implications of these recent regulations are not reflected in the quantitative analysis presented in this report. EPA projects future year decreases in emissions (see Figure 5 in Section 2.2.1). However, better controls are needed for on-road heavy duty diesel vehicles and off-road vehicles, which include locomotives, construction, farm, landscaping equipment, and marine vehicles. For these off-road vehicles, 80-90% NO_x removal is technically achievable (deNevers, 1995; Koebel et al., 2004). Assuming a 40% reduction for these sources, there is a potential reduction of 1.4 Tg. The total reduction for both mobile and stationary sources is then approximately 2 Tg N/yr. Part of achieving such levels of compliance will require the implementation of inspection and maintenance programs or road-side monitoring.

The Committee cautions, however, that achieving such a goal may be inadequate for many areas to meet the new 60 to 70 ppb ozone standard recommended by the EPA Clean Air Scientific Advisory Committee (CASAC) (U.S. EPA CASAC, 2008) or even the 75 ppb standard currently promulgated. Additional measures such as increasing the role of solar- and wind-generated electricity, wider use of hybrid and electric cars, and public transit conducive to energy conservation and reduced emissions should be promoted.

> **Management Goal 1.** *The Committee estimates that if EPA were to expand its NO_x control efforts for emissions of mobile sources and power plants and include implementation of year round controls on stationary sources to protect welfare and avoid adverse effects on ecosystems, a 2.0 Tg N/yr decrease in the generation of reactive nitrogen could be achieved. It is believed that coal-fired utilities could experience a reduction of 0.6 Tg N/yr. Since 2002, emissions have already been reduced by at least 0.3 Tg N/yr; hence, this represents an additional 0.3 Tg N/yr. Approximately 3.4 Tg N/yr can be attributed to mobile sources (highway, off-highway). Assuming a conservative 40% reduction, an additional 1.4 Tg N/yr could be reduced.*

Management Goal 2. Nr discharges and emissions from agricultural lands and landscapes

Section 5.3.4 of this report reviews the various methods that can be used to improve Nr management in agricultural systems. The Committee estimates that crop N-uptake efficiencies can be increased by up to 25% over current levels through a combination of knowledge-based practices and advances in fertilizer technology (such as controlled release and inhibition of nitrification). Crop output can be increased while reducing total Nr by up to 20% of applied synthetic fertilizers, approximately 2.4 Tg N/yr below current levels of Nr additions to the environment. These are appropriate management goals with today's available technologies. Further progress is possible through expanded research programs.

The Committee is concerned about current policies and practices governing biofuel development. Acreage devoted to corn production has increased substantially for corn based ethanol production during the past several years (with nearly one-third of the crop currently devoted to bioethanol production), with fertilizer nitrogen use on corn increasing by at least 10% (an additional 0.5 Tg N/yr), largely to meet biofuel feedstock crop demand. In the absence of Nr controls and a failure to implement best practices, current biofuels policies will make it extremely difficult to reduce Nr transfers to soils, water and air (Simpson et al., 2008). Integrated management strategies will be required.

The Committee also notes with concern the increase of N_2O in the atmosphere. The Committee believes that GHG emissions trading will provide both opportunities and challenges for mitigating Nr environmental and health impacts. Policies and regulations should consider how to reward reductions of N-related GHG. Biofuel subsidies that accurately account for Nr contributions to GHG emissions, certification of individual biofuel plants for GHG impact, and rewards for farmers who reduce N_2O emissions are examples of how an integrated strategy can reduce agricultural GHG impacts. For additional production of liquid biofuels beyond the grandfathered amount in the Energy Independence and Security Act (EISA), EPA has the power to exercise some controls on N_2O emissions through the life cycle GHG accounting requirements. It is the opinion of the Committee that Section 204 of the EISA calling on the Agency to adopt a life cycle approach to the assessment of future renewable fuel standards is a positive step toward a comprehensive analysis.

Section 5.3.4 of this report reviews methods of controlling Nr from landscape runoff through the use of natural or restored wetlands, urban areas, and through the use of best management practices. The Committee finds that flows of Nr into streams, rivers, and coastal systems can be reduced by approximately 20% (~1 Tg N/yr) through improved methods of landscape management and without undue disruption to human commercial and aesthetic activities.

> **Management Goal 2.** *The Committee estimates that crop N-uptake efficiencies can be increased by up to 25% over current practices through a combination of knowledge-based practices and advances in fertilizer technology amounting to ~2.4 Tg N/yr below current amounts of Nr additions to the environment. The Committee further estimates that excess flows of Nr into streams, rivers, and coastal systems can be decreased by approximately 20% (~1 Tg N/yr) through improved landscape management and without undue disruption to agricultural production.*

Management Goal 3. Ammonia emissions from livestock management and manure handling

In spite of gains made over the last several decades in decreasing the amount of NO_x emitted from stationary and mobile combustion sources, the total amount of Nr released into the atmosphere has remained relatively constant. This is related to the essentially unregulated release of ammonia from livestock operations.
As discussed in Section 2.2.3, at the present time, fewer livestock are required to produce more animal products than in the past. For example, since 1975 milk production has increased linearly at the rate of ~ 180 kg milk per cow /yr while milk cow herd population decreased at the rate of ~69,000 head per yr (i.e., the 60% greater amount of milk produced in 2006 compared to 1970 required 25% fewer cows). Animal inventories declined by 10% for beef brood cows from 36 million head in 1970 to 33 million head in 2006, and the inventory of breeder pigs and market hogs declined 8% from 673 million head to 625 million head in the same period, even with similar or greater annual meat production. These trends resulted from greater growth rates of animals producing more meat in a shorter amount of time. In 1970, broilers were slaughtered after 80 days on feed at 1.7 kg live weight, but by 2006 the average weight was 2.5 kg after only 44 days on feed. These trends in requiring fewer animals to produce more animal food products through improved diet and increased production efficiency will continue.

Implementation of improved methods of livestock management and manure handling and treatment to decrease NH_3 emissions that have been developed since 1990 will further decrease ammonia and other gases and odor emissions. For example, sawdust litter helps decrease NH_3 emissions from pig manure with 44-74% of manure N converted to N_2. Storage covers for slurry storage tanks, anaerobic lagoons, and earthen slurry pits decrease emissions from those containments. Anaerobic digestion in closed containment has been studied for many types of applications. Recent research demonstrates reduction in NH_3 emissions after a permeable cover was installed (e.g., a polyethylene cover decreased NH_3 emissions by ~80%). A well managed swine lagoon can denitrify approximately 50% of the excreted N to N_2. Recently engineered developments utilizing closed loop systems (Aneja et al., 2008a) substantially reduce atmospheric emissions of ammonia (> 95%) and odor at hog facilities. Based upon recently demonstrated reduction of NH_3 emissions from swine and poultry production, a moderate reduction of 50% from 1990 NH_3 emission estimates for swine and poultry production should be attainable (Table 17). Because of the larger land area involved in dairy and beef production and the lack of effort that has been exerted in mitigating NH_3 emissions, a more modest and reachable goal of decreasing NH_3 emissions by 10% through improvements in animal diet and manure management is proposed (Table 17).

> **Management Goal 3.** *The Committee estimates that livestock-derived NH_3 emissions can be decreased by 30% (a decrease of 0.5 Tg N/yr) by a combination of BMPs and engineered solutions. This is expected to decrease $PM_{2.5}$ by ~0.3 $\mu g/m^3$ (2.5%), and improve health of ecosystems by achieving progress towards critical load recommendations. Additionally we estimate that NH_3 emissions derived from fertilizer applications can be decreased by 20% (a decrease*

Table 17: Estimates for potential decreases in NH_3 emissions from livestock manure in the United States

NH_3 Source	% of Total NH_3	Tg NH_3-N/yr emitted	Estimated Decrease of NH_3	
			%	Tg N/yr
Dairy	23.1	0.37	10	0.040
Beef	27.1	0.44	10	0.040
Poultry	27.5	0.44	50	0.220
Swine	17.5	0.28	50	0.140
Goat/sheep	1.6	0.03	10	0.003
Horse	2.9	0.05	10	0.005
Total	**100**	**1.61**		**0.45**

Estimate is based on livestock emissions of 1.6 Tg from Table 1.

of ~0.2 Tg N/yr), through BMPs that focus on improvements related to application rate, timing, and placement.

Management Goal 4. Discharge of Nr from developed lands and point sources

National loadings of Nr to the environment from public and private wastewater point sources are relatively modest in comparison with global Nr releases; however, they can be important local sources with associated impacts, especially in highly-populated watersheds. The Committee has estimated that sewage containing Nr from human waste contributes 1.3 Tg N/yr to the terrestrial inputs of nitrogen (Table 1).

The Committee has also estimated that turf fertilizer usage contributes 1.1 Tg N/yr to terrestrial inputs, a load that could potentially be cut by about one third (Section 2.2.4). The Committee did not provide estimates for general stormwater and nonpoint source runoff nitrogen load reductions specific to developed or urban areas – runoff concentrations and loads are highly variable reflecting geographic and climatic conditions throughout the U.S. and equally variable removal efficiencies from standard treatment BMPs. This is shown in a summary of the International Stormwater Best Management Practices Database (Geosyntec Consultants, Wright Water Engineers, Inc., 2008). However, most BMPs are effective because they provide the beneficial biochemical conditions of wetlands, the biophysical controls described in Section 5.3.4 and Appendix C. These benefits, and the application of BMPs, are recommended in overarching recommendation 4, as well as in the preceding Management Goal 2 as applied to agricultural lands. Similar stormwater and nonpoint source management benefits specific to developed lands should be anticipated with BMP application in those areas.

Denitrification processes as applied to human waste at sewage treatment plants are well-studied and growing in application. Performance of these engineered solutions, collectively referenced as biological nitrogen removal (BNR), can be more rigorously controlled than for stormwater and nonpoint source BMPs. Recent publications by the U.S EPA (2007f, 2008e,f) have summarized the state of and the capability for nitrogen removal, and have reported that technologies to achieve effluent concentrations of 3 mg total nitrogen per liter (TN/L) or less are readily available. However, plant capacity and design, wastewater characteristics, and climate conditions can all affect the ability of a facility to remove nitrogen. EPA's review of 2003-2006 data for 16 facilities that remove nitrogen to varying degrees found a range of final effluent TN concentrations of 1.0 to 9.7 mg/L, with an average of 5.6 mg/L. In general, very small facilities (<0.1 MGD) do not perform as well, with a final TN concentration ranging from 6-12 mg/L. Treatment performance varied and exceeded 5 mg TN/L at some of the facilities. Given these conditions, and performance uncertainties, it seems reasonable to conclude that removal efficiencies in the range of 40-60% below standard effluent nitrogen loads could be readily attained. Based on the human waste load of 1.3 Tg N/yr, this would yield a decrease in total nitrogen load of between 0.5 and 0.8 Tg/yr. Using data provided by Maryland Department of the Environment (2006) and the Connecticut Department of Environmental Protection (2007), two states that have promoted nitrogen removal technologies as solutions to coastal eutrophication, EPA (2007f) has constructed cost estimates of upgrading the performance of sewage treatment plants ranging from $990,000 to $1.74 million per MGD treated.

There are two funding sources of significance authorized in the CWA that are used to fund projects relevant to the control of Nr. Section 319 establishes state nonpoint source management programs to plan for and implement management measures that abate sources of nonpoint pollution from eight source categories, including both urban and agricultural sources; however, the CWA disallows use of 319 funds for NPDES permit requirements, so urban areas with stormwater permits do not qualify for Section 319 funding. Over the years, section 319 has made available, through 60% matching funds, over $1.6 billion in assistance. The much larger source of funding comes under Title VI of the CWA, which has provided over $24 billion (federal) for the construction of treatment facilities for point sources of wastewater over the past 20 years, although only a fraction of this amount has been dedicated to denitrification processes. Title VI “state revolving” loan funds can be used for stormwater management, as well as other water pollution management activities, but not all states have chosen to use funds beyond traditional sewage treatment plant infrastructure needs because of the large backlog of demand for those purposes.

In 2009, under the American Reinvestment and Recovery Act (ARRA), the CWA Clean Water State Revolving Fund (CWSRF) received a $4 billion boost for clean water infrastructure and the CWSRF for fiscal year 2010 was tripled over the prior years to two billion dollars. These additional funds not only provide for jobs creation, as intended by Congress, but provide states with resources to reduce the backlog of clean water projects, which also often include nutrient management needs. The ARRA funds also emphasized the use of CWSRF dollars for stormwater and nonpoint source management and energy savings under a “green infrastructure” requirement. A 20% set-aside for green infrastructure was a requirement of AARA CWSRF funding and was used widely for projects that included reductions in GHG emissions, land-based low impact development BMPs to reduce runoff and improve runoff quality, and other innovative practices to treat wastewater and runoff. A green infrastructure requirement is being continued in the fiscal year 2010 CWSRF allocation.

> **Management Goal 4.** *The Committee recommends that a high priority be assigned to increasing funding for nutrient management. We estimate that a decrease in Nr emissions from human sewage of between 0.5 and 0.8 Tg N/yr can be achieved, with additional decreases likely with increased stormwater and nonpoint source BMP application support.*

6.4. Summary of Specific Findings and Recommendations Corresponding to the Four Study Objectives

In this report the Committee has provided specific findings and recommendations to assist EPA in its understanding and management of nitrogen-related air, water, and soil pollution issues. The specific findings and recommendations corresponding to each of the four study objectives are summarized below.

Study Objective #1: Identify and analyze, from a scientific perspective, the problems Nr presents in the environment and the links among them.

In general, the Committee finds that uncertainty associated with rapid expansion of biofuels, losses of Nr from grasslands, forests, and urban areas, and the rate and extent of denitrification have created the need to measure, model, and report all forms of Nr consistently and accurately. Addressing this need will decrease uncertainty in the understanding of the fate of Nr that is introduced into the environment and lead to a better understanding of the impacts of excess Nr on the health of people and ecosystems. This should be accomplished through a coordinated effort among cognizant federal and state agencies and universities.

The Committee recommends that EPA routinely and consistently account for the presence of Nr in the environment in forms appropriate to the medium in which they occur (air, land, and water) and that accounting documents be produced and published periodically.

Specific Findings:

- Rapid expansion of corn-ethanol production has the potential to increase N fertilizer use through expanding corn production and its associated N fertilizer inputs. Development of cellulosic ethanol industry will require cultivation for cellulosic crops, which will also require N fertilizer. Distillers grains are changing animal diets and affecting N recycling in livestock. Both have important consequences for the effective future management of Nr. (Finding #4 – also pertains to study objectives 2 and 4)
- Although total N budgets within all terrestrial systems are highly uncertain, Nr losses from grasslands and forests (vegetated) and urban (populated) portions of the N cascade appear to be higher, on a percent of input basis, than from agricultural lands. The relative amount of these losses ascribed to leaching, runoff, and denitrification are as uncertain as the N budgets themselves. (Finding #9)
- Denitrification of Nr in terrestrial and aquatic systems is one of the most uncertain parts of the nitrogen cycle. Denitrification is generally considered to be a dominant N loss pathway in both terrestrial and aquatic systems, but it is poorly quantified. (Finding #10 – also pertains to study objective 4)
- The Committee finds that there is a need to measure, compute, and report the total amount of Nr present in impacted systems in appropriate units. Because what is measured influences what we are able to perceive and respond to, in the case of Nr, it is especially critical to measure total amounts and different chemical forms at regular intervals over time. (Finding #13 – also pertains to study objective 4)

Specific Recommendations:

- The Committee recommends that EPA routinely and consistently account for the presence of Nr in the environment in forms appropriate to the medium in which they occur (air, land, and water) and that accounting documents be produced and published periodically (for example, in a fashion similar to National Atmospheric Deposition Program summary reports). The Committee understands that such an undertaking will require substantial resources, and encourages the Agency to develop and strengthen partnerships with appropriate federal and state agencies and private sector organizations with parallel interests in advancing the necessary underlying science of Nr creation, transport and transformation, impacts, and management. (Recommendation #13 – also pertains to study objective 4)
- EPA should work with USDA and universities to improve understanding and prediction of how expansion of biofuel production, as mandated by the 2007 EISA, will affect Nr inputs and outputs from agriculture and livestock systems. Rapid expansion of biofuel production has the potential to increase N fertilizer use through expansion of corn production area and associated N fertilizer inputs, and from extending cultivation of cellulosic materials that will also need N inputs. Current models and understanding are not adequate to guide policy on how to minimize impact of biofuel expansion on environmental concerns related to Nr. (Recommendation #4)
- EPA should join with USDA, DOE, and universities in efforts to ensure that the N budgets of terrestrial systems are properly quantified and that the magnitudes of at least the major loss vectors are known. (Recommendation #9 – also pertains to study objectives 2 and 4)
- EPA, USDA, DOE, and universities should work together to ensure that denitrification in soils and aquatic systems is properly quantified, by funding appropriate research. (Recommendation #10 – also pertains to study objective 4)

Study Objective #2: Evaluate the contribution an integrated N management strategy could make to environmental protection.

In general, the Committee finds that effective management of Nr in the environment must recognize the existence of tradeoffs across a number of impact categories involving the cycling of nitrogen and other elements. In addition, an integrated multi-media approach to monitoring Nr is needed.

The Committee recommends that EPA:

1. Develop a uniform assessment and management framework that considers the effects of Nr loading over a range of scales reflecting ecosystem, watershed, and regional levels. The framework should include all inputs related to atmospheric and riverine delivery of Nr to estuaries, their comprehensive effects on marine eutrophication dynamics and their potential for management
2. Examine the full range of traditional and ecosystem response categories, including economic and ecosystem services, as a basis for expressing Nr impacts in the environment, and for building better understanding and support for integrated management efforts.

Specific Findings:

- There has been a growing recognition of eutrophication as a serious problem in aquatic systems (NRC, 2000). The last comprehensive National Coastal Condition Report was published in 2004 (EPA, 2004) and included an overall rating of "fair" for estuaries, including the Great Lakes, based on evaluation of over 2000 sites. The water quality index, which incorporates nutrient effects primarily as chlorophyll-*a* and dissolved oxygen impacts, was also rated "fair" nationally. Forty percent of the sites were rated "good" for overall water quality, while 11% were "poor" and 49% "fair." (Finding #11)
- The Committee finds that reliance on only one approach for categorizing the measurement of Nr is unlikely to result in the desired outcome of translating N-induced degradation into the level of understanding needed to develop support for implementing effective Nr management strategies. (Finding #14)
- Effective management of Nr in the environment must recognize the existence of tradeoffs across impact categories involving the cycling of other elements, particularly carbon and phosphorus. (Finding #18)
- The Committee has determined that an integrated approach to monitoring that includes multiple media (air, land, and water) components and considers a suite of environmental and human concerns related to reactive nitrogen in the environment (e.g., Nr effects, climate change, human health) is needed. Some of the phenomena that presented in this report need more definition and verification. More importantly, however, as controls are brought to bear on Nr, improvements need to be measured to verify and validate successful management strategies. If the desired improvements are not realized as shown by the collected data, corrective measures will be required. Such an adaptive approach acknowledges the likelihood that management programs will be altered as scientific and management understanding improve. (Finding #20 – also pertains to study objective 3)

Specific Recommendations:

- The Committee recommends that EPA develop a uniform assessment and management framework that considers the effects of Nr loading over a range of scales reflecting ecosystem, watershed, and regional levels. The framework should include all inputs related to atmospheric and riverine delivery of Nr to estuaries, their comprehensive effects on marine eutrophication dynamics, and their potential for management. (Recommendation #11)

- It is recommended that the EPA examine the full range of traditional and ecosystem response categories, including economic and ecosystem services, as a basis for expressing Nr impacts in the environment, and for building better understanding and support for integrated management efforts. (Recommendation #14)
- The Committee recommends that the integrated strategies for Nr management outlined in this report be developed in cognizance of the tradeoffs associated with reactive nitrogen in the environment, consistent with the systems approach of overarching recommendations 2 and 3. Specific actions should include:
    - Establishing a framework for the integrated management of carbon and reactive nitrogen
    - Implementing a research program that addresses the impacts of tradeoffs associated with management strategies for carbon, reactive nitrogen, and other contaminants of concern
    - Implementing a research and monitoring program aimed at developing an understanding of the combined impacts of different nitrogen management strategies on the interchange of reactive nitrogen across environmental media. (Recommendation #18)
- In cooperation with the Department of Agriculture, U.S. Army Corps of Engineers, U.S. Fish and Wildlife Service, and the and the Federal Emergency Management Agency, the EPA should develop programs to encourage wetland restoration and creation, with strategic placement of these wetlands where reactive nitrogen is highest in ditches, streams, and rivers. The Agency should also address the means of financing, governance, monitoring, and verification. Such programs might be modeled on the Conservation Reserve Program or extant water quality and environmental trading programs, but need not be limited to current practices. (Recommendation #15e – also pertains to study objective 3)
- The Committee recommends that EPA initiate discussions and take action to develop a national, multi-media monitoring program that monitors sources, transport and transition, effects using indicators where possible, and sinks of Nr in keeping with the nitrogen cascade concept. This comprehensive program should build upon existing EPA and state initiatives, as well as monitoring networks already underway in other federal agencies such as the U.S. Geological Survey programs and the NADP effort. (Recommendation #20 – also pertains to study objective 3)

Study Objective #3: Identify additional risk management options for EPA's consideration.

In general, the Committee finds that a number of risk management actions should be considered to reduce Nr loading and transfer to the environment. The Committee recommends risk management actions that include farm-level improvements in manure management, actions to reduce atmospheric emissions of Nr, and interventions to control Nr in water management programs.

Specific Findings:

- Farm-level improvements in manure management can substantially reduce Nr load and transfer. There are currently very few incentives or regulations to decrease these transfer and loads, despite the existence of management options to mitigate. (Finding #6)
- Scientific uncertainty about the origins, transport, chemistry, sinks, and export of Nr remains high, but evidence is strong that atmospheric deposition of Nr to the earth's surface, as well as emissions from the surface to the atmosphere, contribute substantially to environmental and health problems. Nitrogen dioxide, NO_2, is often a small component of NO_y, the total of oxidized nitrogen in the atmosphere. The current NAAQS for NO_2, as an indicator of the criteria pollutant "oxides of nitrogen," is inadequate to protect health and welfare. NO_y should be considered seriously as a supplement or replacement for the NO_2 standard and in monitoring. Atmospheric emissions and concentrations of Nr from agricultural practices (primarily in the form of NH_3) have not been well monitored, but NH_4^+ ion concentration and wet deposition (as determined by NADP and NTN) appear to be increasing, suggesting that NH_3 emissions are increasing. Both wet and dry deposition contribute substantially to NH_x removal, but only wet deposition is known with much scientific certainty. Thus, consideration should be given to adding these chemically reduced and organic forms of Nr to the list of criteria pollutants. (Finding #8)
- Meeting Nr management goals for estuaries, when a balance should be struck between economic, societal, and environmental needs, seems unlikely under current federal law. Enforceable authorities over nonpoint source, stormwater, air (in terms of critical loads), and land use are not adequate to support necessary Nr controls. Funding programs are presently inadequate to meet existing pollution control needs. Furthermore, new technologies and management approaches are required to meet ambitious Nr control needs aimed at restoring national water quality. (Finding #12 – also pertains to study objective 4)
- Intervention to control Nr under most water management programs generally occurs in three ways:
    - Prevention or source controls
    - Physical, chemical or biological "dead ending" or storage within landscape compartments where it

is rendered less harmful (e.g., long-term storage in soils or vegetation; denitrification, primarily in wetlands; reuse)

  - Treatment using engineered systems such as wastewater treatment plants or BMPs for stormwater and nonpoint source runoff

While most management programs focus on the third (treatment) approach, there are opportunities for combining the three that can be more effective and cost less. (Finding #15)

- The Committee finds that there have been persistent increases in the amounts of Nr that have been emitted into and retained within various ecosystems, affecting their functioning. Unless this trend is reversed, it will become increasingly difficult for many of these ecosystems to provide the services upon which human well-being depends. The Committee believes that there is a need to regulate certain forms of Nr to address specific problems related to excess Nr, and we believe that the best approach for an overall management strategy is the concept of defining acceptable total Nr critical loads for a given environmental system. (Finding #16 – also pertains to study objective 4)
- Current EPA policy (40 CFR Part 51, Clean Air Fine Particle Implementation Rule) discourages states from controlling ammonia emissions as part of their plan for reducing $PM_{2.5}$ concentrations. In this rulemaking (*Federal Register* 72(79): 20586-20667), EPA has stated that "ammonia reductions may be effective and appropriate for reducing $PM_{2.5}$ concentrations in selected locations, but in other locations such reductions may lead to minimal reductions in $PM_{2.5}$ concentrations and increased atmospheric acidity." Ammonia is a substantial component of $PM_{2.5}$ in most polluted areas of the U.S. at most times. While it is true that reducing NH_3 emissions might increase the acidity of aerosols and precipitation, the net effect of NH_3 on aquatic and terrestrial ecosystems is to increase acidity. After being deposited onto the earth's surface, NH_4^+ is, under most circumstances, quickly nitrified, increasing the acidity of soils and waters. The Committee is unaware of any evidence that NH_3 reduces the toxicity of atmospheric aerosols or that high concentrations of NH_3 occur naturally over any substantial area of the U.S. It has not yet been established which components of PM have substantive effects on human health but the total concentration of $PM_{2.5}$ correlates with morbidity and mortality, and NH_3 contributes to $PM_{2.5}$. The visibility, degradation, and other adverse effects associated with $PM_{2.5}$ are related to aerosol surface area or mass where NH_4^+ certainly plays a role. (Finding #17)

Specific Recommendations:

- A policy, regulatory, and incentive framework is needed and should be developed to improve manure management to reduce Nr load and ammonia transfer, taking into account phosphorus load issues. (Recommendation #6)
- EPA should re-examine the criteria pollutant "oxides of nitrogen" and the indicator species, NO_2, and consider adding chemically reactive nitrogen as a criteria pollutant and NH_x and NO_y as indicators to supplement the NO_2 National Ambient Air Quality Standards. (Recommendation #8a)
- The Committee recommends that monitoring of NH_x and NO_y begin as soon as possible to supplement the existing network of NO_2 compliance monitors. (Recommendation #8b)
- The Committee recommends that EPA reevaluate water quality management approaches, tools, and authorities to ensure Nr management goals are attainable, enforceable, and the most cost-effective available. Monitoring and research programs should be adapted as necessary to ensure they are responsive to problem definition and resolution, particularly in the development and enhancement of nitrogen removal technologies and best management practices, and continue to build our level of understanding and increase our ability to meet management goals. (Recommendation #12 – also pertains to study objective 4)
- To better address Nr runoff and discharges from the peopled landscape, the Committee recommends that EPA:
  - Evaluate the suite of regulatory and non-regulatory tools used to manage Nr in populated areas from nonpoint sources, stormwater and domestic sewage, and industrial wastewater treatment facilities, including goal-setting through water quality standards and criteria; and
  - Determine the most effective regulatory and voluntary mechanisms to apply to each source type with special attention to the need to regulate nonpoint source and related land use practices. (Recommendation #15a)
  - Review current regulatory practices for point sources, including both wastewater treatment plants and stormwater, to determine adequacy and capacity towards meeting national Nr management goals; and
  - Consider technology limitations, multiple pollutant benefits, and funding mechanisms as well as potential impacts on climate change from energy use and greenhouse gas emissions, including nitrous oxide. (Recommendation #15b)
  - Set Nr management goals on a regional/local basis, as appropriate, to ensure most effective use of limited management dollars; and
  - Fully consider "green" management practices such as low-impact development and conservation measures

that preserve or re-establish Nr removing features to the landscape as part of an integrated management strategy along with traditional engineered best management practices. (Recommendation #15c)

- The Committee recommends that the Agency work toward adopting the critical-loads approach concept in determining thresholds for effects of excess Nr on terrestrial and aquatic ecosystems. In carrying out this recommendation, the Committee recognizes that in many cases it will be necessary for the Agency to enter into new types of research, policy, and regulatory agreements with other federal, state, and tribal units based on cooperative, adaptive, and systemic approaches that derive from a common understanding of the nitrogen cascade. (Recommendation #16 – also pertains to study objective 4)
- The Committee recommends that the EPA presumption that NH_3 is not a $PM_{2.5}$ precursor should be reversed and states should be encouraged to address NH_3 as a harmful $PM_{2.5}$ precursor. (Recommendation #17)

Study Objective #4: Make recommendations to EPA concerning improvements in Nr research to support risk reduction.

The Committee has recommended research in key areas to support risk reduction. The Committee's recommendations include research to advance the understanding of: the quantity and fate of Nr applied to major crops; how to accelerate crop yields while increasing N fertilizer uptake efficiency; agricultural emissions of forms of Nr; atmospheric deposition of Nr; and the potential for amplification of Nr-related climate impacts.

Specific Findings:

- Crop agriculture receives 60% of U.S. annual new Nr inputs from anthropogenic sources (9.8 Tg from N fertilizer, 7.7 Tg from crop BNF versus 29 Tg of N anthropogenically introduced into the U.S. environment per year) and accounts for 58% (7.6 Tg) of total U.S. Nr losses from terrestrial systems to air and aquatic ecosystems, yet current monitoring of fertilizer use statistics by federal agencies is inadequate to accurately track trends in quantities and fate of N applied to major crops and the geospatial pattern by major watersheds. (Finding #1)
- Nr inputs to crop systems are critical to sustain crop productivity and soil quality. Moreover, given limited land and water resources, global population growth, and rapid economic development in the world's most populous countries, the challenge is to accelerate increases in crop yields on existing farm land while also achieving a substantial increase in N fertilizer uptake efficiency. This process is called "ecological intensification" because it recognizes the need to meet future food, feed, fiber, and energy demand of a growing human population while also protecting environmental quality and ecosystem services for future generations (Cassman, 1999). More diverse cropping systems with decreased Nr fertilizer input may also provide an option on a large scale if the decrease in Nr losses per unit of crop production in these diverse systems can be achieved without a decrease in total food production, which would trigger indirect land use change to replace the lost production and negate the benefits. However, crop cultivars and agronomic practices are changing rapidly, which changes N requirements, but current efforts in research, extension, and conservation programs on N management within these rapidly evolving systems are not adequate to meet the challenge of providing better information to increase NFUE. (Finding #2)
- Nitrous oxide emissions from the Nr inputs to cropland from fertilizer, manure, and legume fixation represent a large proportion of agriculture's contribution to greenhouse gas emissions, and the importance of this source of anthropogenic greenhouse gas will likely increase unless NFUE is markedly improved in crop production systems. Despite its importance, there is considerable uncertainty in the estimates of nitrous oxide emissions from fertilizer, and research should focus on reducing this uncertainty. (Finding #3)
- There are no nationwide monitoring networks in the U.S. to quantify agricultural emissions of greenhouse gases, NO, N_2O, reduced sulfur compounds, VOCs, and NH_3. In contrast, there is a large network in place to assess the changes in the chemical climate of the U.S. associated with fossil fuel energy production, i.e., the National Atmospheric Deposition Program/National Trends Network (NADP/NTN) which has been monitoring the wet deposition of sulfate (SO_4^{2-}), NO_3^-, and NH_4^+ since 1978. (Finding #5)
- Synthetic N fertilizer application to urban gardens and lawns amounts to approximately 10% of the total annual synthetic N fertilizer used in the U.S. Even though this N represents a substantial portion of total N fertilizer use, the efficiency with which it is used receives relatively little attention. (Finding #7)
- The biogeochemical cycle of Nr is linked to climate in profound but nonlinear ways that are, at present, difficult to predict. Nevertheless, the potential for significant amplification of Nr-related impacts is substantial, and should be examined in more complete detail. (Finding #19)

Specific Recommendations:

- The Committee recommends increasing the specificity and regularity of data acquisition for fertilizer application to major agricultural crops in terms of timing and sufficiently small application scale (as well as for urban residential and recreational turf) by county (or watershed) to better inform decision-making about policies and mitigation options for reducing Nr load in these systems, and to facilitate monitoring and evaluation of impact from implemented policies and mitigation efforts. (Recommendation #1)

- To obtain information on Nr inputs and crop productivity the Committee recommends that:
  - Data on NFUE and N mass balance, based on direct measurements from production-scale fields, be generated for the major crops to identify which cropping systems and regions are of greatest concern with regard to mitigation of Nr load, and to better focus research investments, policy development, and prioritization of risk mitigation strategies. (Recommendation #2a)
  - Efforts at USDA and universities be promoted to: (1) investigate means to increase the rate of gain in crop yields on existing farm land while increasing N fertilizer uptake efficiency, and (2) explore the potential for more diverse cropping systems with lower N fertilizer input requirements to the extent that large-scale adoption of such systems would not cause indirect land use change. (Recommendation #2b)
  - EPA work closely with the U.S. Department of Agriculture (USDA), Department of Energy (DOE), the National Science Foundation (NSF), and universities to help identify research and education priorities to support more efficient use and better mitigation of Nr applied to agricultural systems. (Recommendation #2c)
- The Committee recommends that EPA ensure that the uncertainty in estimates of nitrous oxide emissions from crop agriculture be greatly reduced through the conduct of EPA research and through coordination of research efforts more generally with other agencies such as USDA, DOE, NSF, and with research conducted at universities. (Recommendation #3)
- The status and trends of gases and particulate matter emitted from agricultural emissions (e.g., NO_3^- and NH_4^+) should be monitored and assessed utilizing a nationwide network of monitoring stations. EPA should coordinate and inform its regulatory monitoring and management of reactive nitrogen with the multiple efforts of all agencies including those of the U.S. Department of Agriculture and NSF-supported efforts such as the National Ecological Observatory Network (NEON) and the Long Term Ecological Research Network (LTER). (Recommendation #5)
- To ensure that urban fertilizer is used as efficiently as possible, the Committee recommends that EPA work with other agencies such as USDA as well as state and local extension organizations to coordinate research and promote awareness of the issue. (Recommendation #7a)
- Through outreach and education, supported by research, improved turf management practices should be promoted, including improved fertilizer application and formulation technologies and maintenance techniques that minimize supplemental Nr needs and losses, use of alternative turf varieties that require less fertilization, alternative ground covers in place of turf, and use of naturalistic landscaping that focuses on native species. (Recommendation #7b)
- EPA should pursue the longer term goal of monitoring individual components of Nr, such as NO_2 (with specificity), NO, PAN, and HNO_3, and other inorganic and reduced forms, as well as support the development of new measurement and monitoring methods. (Recommendation #8c)
- The scope and spatial coverage of the Nr concentration and flux monitoring networks (such as the National Atmospheric Deposition Program and the Clean Air Status and Trends Network) should be increased, and an oversight review panel should be appointed for these two networks. (Recommendation # 8d)
- EPA in, coordination with other federal agencies, should pursue research goals including:
  - Measurements of deposition directly both at the CASTNET sites and in nearby locations with non-uniform surfaces such as forest edges
  - Improved measurements and models of convective venting of the planetary boundary layer (the lowest layer of the atmosphere) and of long range transport
  - Improved analytical techniques and observations of atmospheric organic N compounds in vapor, particulate, and aqueous phases
  - Increased quality and spatial coverage of measurements of the NH_3 flux to the atmosphere from major sources especially agricultural practices
  - Improved measurement techniques for, and numerical models of, NO_y and NH_x species (especially with regard to chemical transformations, surface deposition and off-shore export and linked ocean-land-atmosphere models of Nr). (Recommendation #8e)
- Research should be conducted on: best management practices that are effective in controlling Nr, especially for nonpoint and stormwater sources (including land and landscape feature preservation); setting Nr management targets that realistically reflect these management and preservation capacities; and constructing a decision framework to assess and determine implementation actions consistent with management goals. (Recommendation 15d)
- The EPA should support cross-disciplinary and multiagency research on the interactions of climate and Nr. To determine the interactions of global biogeochemical Nr cycles and climate, the

Committee suggests that EPA follow a series of steps such as:

1. Select several likely scenarios for global climate from the IPCC report for the year 2050
2. Down-scale statistics or nest regional climate models within each of these global scenarios to generate meteorological and chemical fields (e.g., temperature, relative humidity, winds, precipitation, CO_2) for a few years around 2050
3. Run several independent biogeochemical Nr models (earth system models that include air/water/land) for North America for these years with current Nr and emissions and application rates
4. Rerun models with decreased Nr emissions/application to evaluate strategies for controlling impacts such as those described in this report. (Recommendation #19)

6.5 Conclusions and Observations

Nitrogen is not only an essential resource for humans, for example its use in food production, but also a byproduct of essential processes such as combustion. The increasing amounts of nitrogen that humans capture through the Haber-Bosch process for useful purposes and the increasing amounts of nitrogen released from combustion also add increasing amounts of excess reactive nitrogen (Nr) to the environment. In the United States, the production of crops receives 60% of the new Nr inputs from anthropogenic sources, but also accounts for almost the same proportion of total Nr losses from terrestrial systems to air and aquatic ecosystems. Nitrogen fertilizer use efficiency by crops seldom reaches 50% and may be 33% or lower. Fossil fuel combustion is another major contributor of excess Nr in the production of essential energy. However, the excess Nr that flows from these activities is not benign. There are serious negative impacts from excess Nr on both human health and the environment. Table 1 gives a range of examples of these.

The nature of the problem

Dealing with excessive reactive nitrogen is an extraordinarily complex issue. Part of this relates to the nature of nitrogen and its ability to change its form and flow through different media, as evidenced conceptually by the nitrogen cascade. Nitrogen's transformative nature only increases the difficulty of dealing with its negative aspects. Further, unlike the linear problems that society has been more accustomed to dealing with in the past, excessive Nr in the environment is in a class of problems sometimes characterized as "wicked" (Batie 2008, Kreuter 2004). For a "wicked" problem;

- There is not universal agreement on what the problem is – different stakeholders define it differently
- There is no defined end solution, the end will be assessed as "better" or worse"
- The problem changes over time
- There is no clear stopping rule – stakeholders, political forces and resource availability will make that determination on the basis of "judgments"
- The problem is associated with high uncertainty of both components and outcomes
- Values and societal goals are not necessarily shared by those defining the problem or those attempting to make the problem better

This is not to say that society does not try to deal with such problems (healthcare, environmental degradation, water resource management, food safety, etc.). What it does say is that different approaches are needed to deal with such problems as compared with better defined problems that are amenable to disciplinary linear science where experts can define the problem and its endpoint.

The Integrated Nitrogen Committee has provided a number of findings and recommendations with respect to the problems caused by excess Nr. Some of these respond to specific defined science based concerns. Others relate to broader concerns the Committee raised in dealing with the overall Nr dilemma. The following points synthesize some of the important lessons learned from the more than four years of study on this issue by the Integrated Nitrogen Committee.

Recognizing the problem and building consensus

It is critically important that the problems caused by integrated nitrogen be widely recognized. Many recognize that we add nitrogen to the environment for specific useful purposes. Fewer recognize that there are both direct and indirect impacts from this that damage both the environment and human health. Until there is general recognition that there is a problem and we need to deal with the negative externalities of excess reactive nitrogen in our environment, there will be no willingness to tackle this issue. Education, communication and outreach are critically important to engender in the public sufficient will to tackle this widespread problem. Education, communication and outreach will be critical to the formation of a common definition of the problem, an essential step once it is recognized. Following that, there will have to be some degree of public consensus for actions that will effectively reduce excess reactive nitrogen in the environment. These steps will not be possible unless there is a process of public consensus building with respect to Nr. The first essential step in trying to deal with a "wicked" problem is getting some measure of agreement across different participants and stakeholders about the problem itself.

The importance of the nitrogen cascade

Understanding the problem will require recognition of the nature of Nr. In this case, the Integrated Nitrogen Committee found the concept of the nitrogen cascade (pictured in the Executive Summary and Chapter 1) to be an essential guide to approaching the problem. This conceptual framework traces the flows of new nitrogen through atmospheric, terrestrial, and aquatic environmental systems where Nr is received, stored and passed on in one form or another. The Committee initially spent much of its effort on understanding these flows and then determining as best as possible the magnitudes of the flows and sinks through these systems. This is the critical baseline information needed to understand the problem, and many of the recommendations in this report relate to gaining a better understanding of this phenomenon. Unless it is well understand how Nr flows through these systems, what the Nr sinks are in these system components, and what the magnitudes are of both the sinks and flows, any attempts to better control Nr may well be ineffective. Understanding and being able to quantify the nitrogen cascade will allow the identification of the major excess Nr contributors, an understanding of the different forms of Nr at various points along the cascade, and enable the determination of more effective interdiction points. Without this basic understanding of the flows of Nr, their nature, and their magnitude, attempts to deal with excess Nr will have high uncertainty. Chapter 2 summarizes much of this critical information that the Committee was able to determine and assemble from other sources.

Integrated approaches are essential

Given what we know about the way Nr behaves, efforts to deal with excess Nr must be organized in a way that reflects the nature of the problem. Unfortunately, many of our approaches are narrowly disciplinary focused, and our policy and regulatory institutions are often focused on one or another media where excess Nr may temporarily reside or on a sector that contributes excess Nr. The regulatory structure that has evolved for problems affecting human health and the environment has been specifically narrow, following policymakers focus on such things as clean water or clean air. These silos have to be broken down if excessive Nr is to be dealt with effectively. Current efforts by EPA and other agencies to encourage more integration across these silos, and include other institutions and stakeholders, are absolutely essential. Research efforts to better understand excess Nr and better mitigate its negative impacts must be trans-disciplinary. What will also be critical is greatly increased collaboration and cooperation among and between agencies and interested stakeholders. Several of the Committee's recommendations address this issue, calling for formal mechanisms to encourage this to take place. The Nr problem has boundaries much broader than the boundaries of the institutions that we have relied on in the past to protect human health and the environment. We are not going to be effective if we do not both expand these boundaries and adopt a broad multi-institutional reach.

Essential monitoring and research

In the effort to understand and quantify the nitrogen cascade the Committee became aware of areas of needed research and monitoring. Much of this is essential for improving our knowledge of what goes on in the cascade. It is also essential for benchmarking current Nr flows and sinks and for targeting where actions are best taken to reduce excess Nr. There is not sufficient information and understanding of these flows and sinks to allow maximum benefit from utilizing the full power of the cascade concept. The Committee has recommended improved monitoring and research to enhance our understanding in air, land and water environments. In some cases our knowledge has such wide margins of error that we cannot identify or quantify important concentrations or flows sufficiently for necessary decisions. In other cases we need much better understanding of the efficacy of actions that might be taken to control Nr. In some cases we need to know more about the indirect impacts of Nr as well as the indirect impacts of measures to control Nr. Monitoring is both an essential part of the research needs as well as being a critical guide to what we face and whether our efforts to better control Nr are being effective. Such environmental monitoring is often not considered to be critically important. It is critically important for effective approaches to reducing excess Nr.

Where to begin

The approach taken in this report and its recommendations should result in helping enable the control of excess Nr to proceed on the basis of starting where it is most technically and economically effective. Some of this may be low hanging fruit, or particular niches where institutions and stakeholders already have a common purpose. Part of the value of the nitrogen cascade is that its characterization of Nr sinks and flows allows the comparison of alternative points of entry and interdiction. Further monitoring and research should also allow the comparison of different modes of interdiction. There will be choices to be made between preventing increased Nr at the source, stabilizing it, or treating the medium involved to remove it. Additional research and actual experience in such efforts, coupled with monitoring, will be essential for making these choices. One product of the cascade exercise is a better understanding of what and where the major contributors of Nr are. The big sources of Nr have to be addressed if there is to be meaningful impact on reducing excess Nr. Part of the decision about where to begin will relate to the efficacy of measures to reduce excess Nr, and our knowledge is by no means complete on this. Beyond the technical considerations of effectiveness, there will have to be policy decisions about the trade-offs and interdependencies between approaches such as market mechanisms, regulation, incentives, and voluntary actions. Market mechanisms generally require regulations that are the basis for the creation of the market situation that makes such things as emission trading viable. Voluntary actions are more likely if there is some prospect of future required action.

Metrics matter

What is measured is critical to determining the dimension of the problem, what to tackle, and whether progress is being made. Kilograms removed (or prevented at the source), the percentage of Nr removed from a particular cascade or sink, or dollars of damage avoided by the removal of Nr are all reasonable measures to use. Measuring the improvement in physical units of Nr can lead to very different source targets, control measures and estimations of success as compared to measuring dollars of damage. This relates to the dilemma presented by "wicked" problems in both the original problem definition and the determination of whether actions have made things better or worse. An example from the Chesapeake Bay is cited in the report that illustrates this. If the goal is less Nr in the bay waters, then all sources are important. If one is concerned about reducing the economic damage of Nr and uses dollars of damage, then atmospheric deposition becomes the primary focus because of the high value of health damages from atmospheric pollution related to Nr.

Setting goals for action

Finally, nothing is going to be accomplished if goals are not set and efforts do not get underway. There are sufficient findings and recommendations presented by the Committee in addition to what we already know to enable multiple agencies to begin to reduce the excess Nr entering the environment. Some of the trade-offs between alternative approaches are also well known. The Committee suggests actions that might be taken by EPA or other management authorities to reduce nitrogen in the environment and a 25% reduction of excess Nr is suggested as attainable with current technology over the near term. Actions being suggested need testing, refining and require monitoring. A start, even on a pilot scale, in one portion of the cascade or sector will yield valuable information about the efficacy of the approaches used, further demonstrate the necessity for a multi-agency joint stakeholder approach, and help further define the problem and where it can initially be best addressed. This necessity of getting underway is one of the main recommendations of the National Research Council's series of reports on improving water quality in the Mississippi (National Research Council, 2008b and 2009). These reports also emphasize the institutional arrangements that are necessary. These are also echoed in this report. Following on the National Research Council's reports, the USDA Natural Resources Conservation Service has recently begun an effort that targets nutrient reductions in the Upper Mississippi. It is time to get more efforts underway with effective collaboration between public and private institutions and stakeholders.

Because this report addresses the needs of EPA's research mission, there is substantial emphasis on gaps in knowledge about Nr and the research that needs to be done to fill these gaps. However, the report contains an extensive knowledge base about Nr which was a necessary precursor to addressing the objectives of the report. The report then identifies the problems posed by Nr, assesses the necessity of an integrated strategy to deal with these problems, identifies some risk management options for EPA's consideration, and makes recommendations for improved research and monitoring to support risk reduction. This information is sufficient to allow initial determinations of where and how the Nr problem can be addressed effectively with positive results. As efforts progress, more will be learned and improved methods, targeting, and analysis can be applied to this truly wicked problem.

Appendix A:
Nitrogen Deposition from the Atmosphere to the Earth's Surface

Review of Nr wet deposition

Substantial progress has been made in monitoring Nr wet deposition as is summarized in information provided by the National Atmospheric Deposition Program/ National Trends Network (NADP) established in 1979. This network monitors precipitation composition at over 250 sites in the U.S. and its territories (http://nadp.sws.uiuc.edu). Precipitation at each station is collected weekly according to well established and uniform procedures and sent to the Central Analytical Laboratory for analysis of acidity, NO_3^-, NH_4^+, chloride, as well as the base cations calcium, magnesium, potassium and sodium. For greater temporal resolution, the Atmospheric Integrated Research Monitoring Network AIRMON, composed of seven sites, was formed in 1992 as part of the NADP program to study wet deposition composition and trends using samples collected daily. The same species are measured as in NADP. By interpolating among sites, NADP is able to estimate the wet deposition of NH_4^+ (reduced N), and NO_3^- (oxidized N) for the 48 contiguous states (Table A-1 and Figure A-1).

Table A-1: Annual wet deposition of reduced (NH_4^+), oxidized (NO_3^-), and total N to the 48 contiguous states

NADP/NTN deposition estimates

	reduced N in precipitation, kg/ha/yr	oxidized N in precipitation, kg/ha/yr	total wet N deposition, kg/ha/yr
1994	1.49	1.68	3.17
1995	1.63	1.67	3.30
1996	1.66	1.80	3.45
1997	1.49	1.74	3.24
1998	1.72	1.78	3.49
1999	1.46	1.58	3.04
2000	1.48	1.62	3.10
2001	1.50	1.57	3.07
2002	1.59	1.55	3.14
2003	1.72	1.55	3.27
2004	1.70	1.52	3.22
2005	1.65	1.41	3.06
2006	1.65	1.40	3.05

Source: NADP/National Trends Network (NTN) http://nadp.sws.uiuc.edu

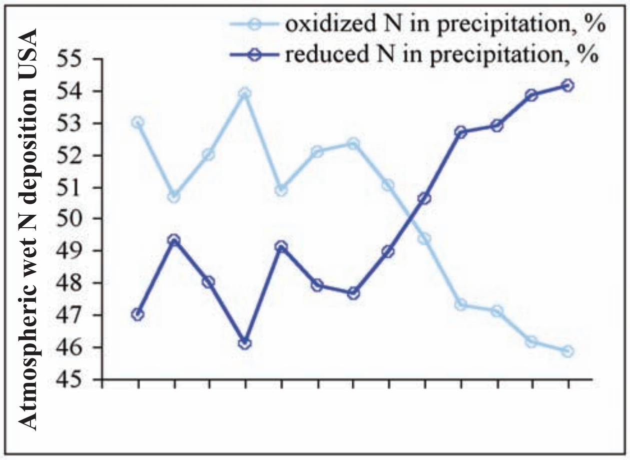

Figure A-1: Percent contribution of oxidized (NO_3^-) and reduced (NH_4^+) nitrogen wet deposition from 1994 to 2006.

As emissions of NO_x have decreased, the relative importance of NH_x has increased (data from National Atmospheric Deposition Program, 2010).

Although individual regions vary, the NADP data for the entire 48 states indicate an apparent decrease in NO_3^- wet deposition, but not in NH_4^+ deposition (Table A-1 and Figure A-2). Ammonium wet deposition shows a weak increase, although the correlation coefficient is small. As NO_x controls have become more effective, the role of reduced N appears to have grown in relative importance. The nitrate data appear to show a statistically significant trend and quantifying the response of deposition to a change in emissions would be useful to both the scientific and policy communities. A notable reduction in power plant NO_x emissions occurred as the result of the NO_x State Implementation Plan (SIP) call (McClenny et al., 2002; Gilliland et al., 2008; Bloomer et al., 2009). EPA should pursue a rigorous analysis of the emissions and deposition data, including identifying monitors and methods that are consistent from the beginning to the end of the record, as indicated in Recommendation 8.

How is Nr deposition related to emissions?

The relationship between emissions of Nr and observed deposition is critical for understanding the efficacy of abatement strategies as well as for partitioning local and large-scale effects of emissions. Only a few studies covering several individual sites have sufficient monitoring consistency and duration to determine rigorously long-term trends in NO_3^- and NH_4^+ and their relationship to emissions, and here we consider several

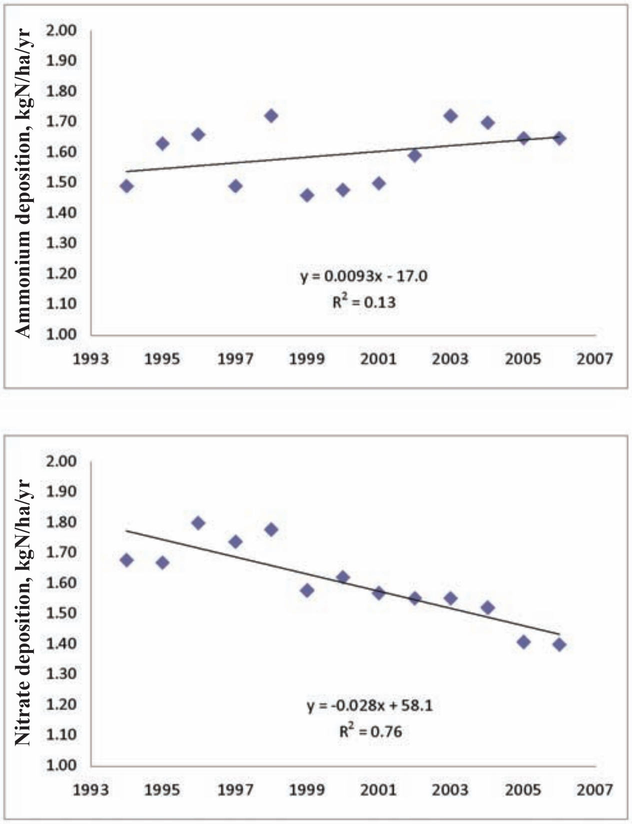

Figure A-2: Trend in reported wet deposition of NH_4^+ and NO_3^- for the 48 contiguous states

Note the sampling methods and locations have not been tested for temporal or spatial bias (data from National Atmospheric Deposition Program, 2010).

examples (Kelly et al., 2002; Butler et al., 2005; Likens et al., 2005). These sites tend to be in the eastern U.S. where monitoring is more concentrated and has a longer history and where upwind sources and downwind receptors are relatively well known. Examination of these studies reveals that concentrations of gaseous and particulate N species in the atmosphere, as well as the Nr content of precipitation over the eastern U.S., shows significant decreases. Correlation with regional emissions is stronger than with local emissions, in keeping with the secondary nature of the major compounds – NO_3^- and NH_4^+. Decreases in NH_4^+ concentration and wet deposition are attributed to decreases in SO_4^{2-} concentrations, meaning that more of the reduced Nr remains in the gas phase. For the period 1965 to 2000, NO_3^- levels in bulk deposition correlate well with reported NO_x emissions. For shorter and earlier time periods the correlation is weaker, and the authors attribute this to changes in the EPA's methods of measuring and reporting emissions; they find evidence of continued errors in emissions from vehicles. Decreases in deposition will probably not be linearly proportional to decreases in emissions; for example a 50% reduction in NO_x emissions is likely to produce a reduction of about 35% in concentration and deposition of nitrate.

The relationship between chemically reduced N emissions and deposition is more complex. The maps of ammonium deposition (Figure A-3) show that maxima occur near or downwind of major agricultural centers where emissions should be high. The full extent of the deposition record (see http://nadp.sws.uiuc.edu) shows the large intensification of NH_4^+ wet deposition in selected areas. The southeastern U.S., particularly North Carolina, has seen a long-term rise (Aneja et al., 2001; Aneja et al., 2003; Stephen and Aneja, 2008). The increase in deposition coincides with the increase in livestock production, but a swine population moratorium appears to have helped abate emissions (Stephen and Aneja, 2008).

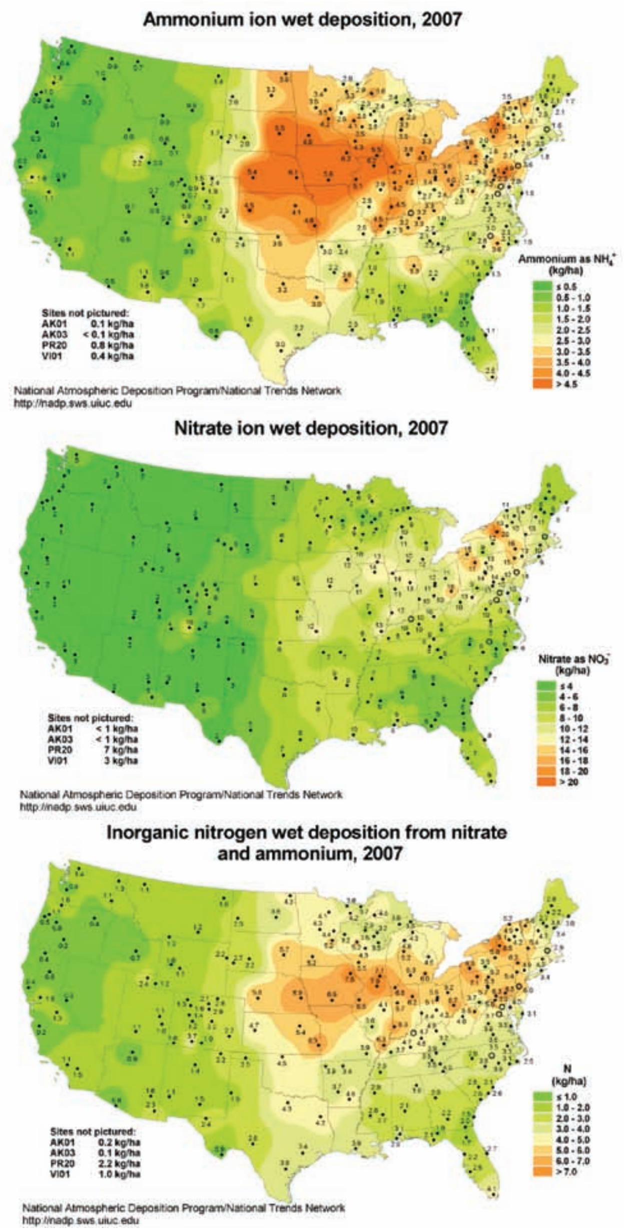

Figure A-3: Annual NH_4^+, NO_3^-, and total inorganic N deposition for the year 2007 showing spatial patterns of deposition

Source: National Atmospheric Deposition Program, 2010.

Concentrations of aerosol NH_4^+ have decreased in many parts of the country, and this may appear to contradict the trend in wet deposition, but a decrease in condensed phase NH_4^+ will be accompanied by an increase in vapor phase NH_3 if SO_4^{2-} and NO_3^- concentrations decrease; see http://vista.cira.colostate.edu/improve/. This potentially misleading information highlights the need for measurements of speciated NH_x (Sutton et al., 2003).

Review of dry deposition observations for the eastern United States

Monitoring dry deposition presents a greater challenge than monitoring wet deposition. The Clean Air Standards and Trends Network (CASTNET) and Atmospheric and Integrated Research Monitoring Network (AIRMON) were established to monitor chemical and meteorological variables to infer dry deposition in order to study the processes leading from emissions to atmospheric concentrations and through deposition to ecosystem effects. AIRMON dry deposition monitoring was discontinued in 2003. See www.epa.gov/castnet/, www.arl.noaa.gov/research/programs/airmon.html, and http://nadp.sws.uiuc.edu.

Recent reviews (Sickles and Shadwick, 2007a,b) analyze the seasonal and regional behavior of concentration and deposition of a variety of primary and secondary pollutants including Nr and investigated trends from 1990 to 2004 for the U.S. east of the Mississippi River. The investigators evaluated observations from more than 50 sites in the eastern states and concluded that for 2000-2004, the mean annual total measured N deposition for this area was 7.75 kg N per hectare per year (expressed as kg N/ha/yr); see Table A-2. This value includes vapor phase HNO_3, particulate NO_3^-, and NH_4^+; it does not include deposition of other oxidized species such as NO_x and PAN, nor gas-phase reduced N species, most notably NH_3. The measured deposition rates peak in spring and summer, but unaccounted for ammonia deposition is probably a substantial fraction of the total, and the true annual cycle remains uncertain.

Table A-2: Deposition of N to the eastern United States in units of kg N/ha/yr

	Annual deposition kg N/ha/yr
Dry NH_4^+	0.41
Wet NH_4^+	2.54
Dry HNO_3 + NO_3^-	1.88
Wet NO_3^-	2.92
Total measured N Dep.	7.75
Est. dry other NO_y	0.94
Est. dry NH_3	1.90
Est. total NO_y	5.74
Est. total NH_3 + NH_4^+	4.85
Est. Grand Total	**10.59**

Data are from the U.S. CASTNET program for the period of 2000-2004. Monitored species for 34 sites east of the Mississippi include vapor-phase HNO_3, particulate NO_3^-, and NH_4^+; unmonitored are other oxidized species such as NO_x and PAN and gas-phase reduced N species most notably NH_3 (Sickles and Shadwick, 2007a). For an explanation of how deposition of unmeasured species was estimated see text.

Estimated total N deposition to the eastern United States

CASTNET monitors HNO_3 and NO_3^-, but not other members of the NO_y family – notably NO_x. Dennis (U.S. EPA, 2007d) estimated that the unmeasured NO_y species account for about 50% of the dry deposition of nitrates. Half of 1.88 (see Table A-2) is 0.94 kg N /ha/yr. Ammonia is also unmeasured by CASTNET, and model estimates (Mathur and Dennis, 2003) of NH_3 indicate that dry deposition should account for 75% of wet NH_4^+ deposition; 75% of 2.54 is 1.9 kg N /ha/yr. Adding these two values to the total from Table A-2 yields a reasonable estimate, within about ±50% absolute accuracy, of total deposition of about 10.6 kg N /ha/yr for the eastern U.S.

Characteristics of N deposition to the eastern United States

Analysis of production of N_2 and N_2O via gas phase reaction is provided in Appendix E. Warmer temperatures are conducive to release of NH_3 from soils and manure as well as from atmospheric particles, thus ammonia concentrations are typically highest in summer. Diffusion of gases is faster than diffusion of particles, and dry deposition of vapor-phase Nr is faster as well; for example the mean CASTNET reported HNO_3 deposition velocity is 1.24 cm/s while that for particulate NO_3^- is 0.10 cm/s. In 2003 and 2004 substantial reductions in emissions from electricity-generating units (power plants) were implemented under the NO_x State Implementation Plan (SIP) call. Many of these power plants are located along the Ohio River generally upwind of the measurement area. Significant reductions ($p = 0.05$) were found between the 1990-1994 and 2000-2004 periods (Sickles and Shadwick, 2007a).

Uncertainty in measured deposition

Analysis of uncertainties in the deposition of Nr is challenging. The coefficient of variation for total regional N deposition for 2000-2004 is 23%, representing a minimal value of uncertainty. Concentrations of some of the NO_y species are monitored, as is the wet deposition of major oxidized and reduced N species, but concentrations of ammonia and other Nr species are not monitored. The network for monitoring dry deposition is sparse and has not been evaluated for spatial bias. The monitors are located in flat areas with uniform surfaces – advective deposition into, for example, the edges of forests are estimated to contribute substantially to the uncertainty (Hicks, 2006). Other sources of error include the model used to convert weekly average concentrations and micrometeorological measurements into depositions. Precision can be determined from collocated sites and is estimated at 5% for nitrate and 15% for ammonium in precipitation (Nilles et al., 1994). The uncertainty in estimated dry deposition arises primarily from uncertainty in deposition velocities (Hicks et al., 1991; Brook et al., 1997) and can be as high as 40% for HNO_3. Total uncertainty for deposition of Nr based on measurements is at least 25% and may be as high as 50%.

Deposition estimates from numerical models

The EPA Community Multiscale Air Quality model (CMAQ) was run for North America at 36 km resolution (R. Dennis et al., personal communication, January 2008). Simulation of Nr deposition is hampered by the lack of emissions information (especially for NH_3), by the need to parameterize planetary boundary layer (PBL) dynamics and deep convection, as well as by simplified multiphase chemistry. This run of CMAQ did not account for NO_x emissions from marine vessels, and these amount to about 4% of the total NO_x emissions in 2000. Calculated nitrogen deposition for the 48 contiguous states (Table A-3) was broadly consistent with direct measurements (Table A-2). CMAQ NO_x emissions were 5.84 Tg N for the year 2002; of that, 2.74 Tg N were deposited. This suggests that ~50% was exported – a number somewhat higher than has been reported in the literature; this discrepancy is discussed below.

Table A-3: Results from CMAQ for total deposition in 2002 to the 48 contiguous states of oxidized and reduced N*

	kg N/ha/yr	Tg N/yr
Oxidized N	3.51	2.74
Reduced N	2.66	2.07
Total N Deposition	6.17	4.81

The CMAQ results were adapted from Schwede et al., (2009) The Watershed Deposition Tool: A tool for incorporating atmospheric deposition in water-quality analyses, htttp://www.epa.gov/amad/EcoExposure/depositionMapping.html.

Ammonia emissions and ambient concentrations can be measured, but are not routinely monitored. For Nr, the CMAQ numerical simulation employed inverse modeling techniques – that is, NH_3 emissions were derived from observed NH_4^+ wet deposition (Gilliland et al., 2003; Mathur and Dennis, 2003; Gilliland et al., 2006). Model determinations of NH_3 therefore do not provide an independent source of information on NH_4^+ deposition.

The three-year CMAQ run gives an indication of the spatial pattern of deposition (Figures A-4). For NH_x, wet and dry are equally important, but for NO_y, dry deposition accounts for about two-thirds of the total deposition, while wet deposition accounts for about one-third. While this does not hold for the eastern U.S. it is true for the U.S. as a whole; in arid southern California, for example, dry deposition of Nr dominates. Based on CMAQ, total NO_y deposition is 2.79 times the wet deposition and total NH_x deposition is 1.98 times the wet deposition. Using the data from Table A-1 for the average wet deposition for the period 2000- 2004, total deposition of oxidized N is 4.36 kg N /ha/yr (2.79 × 1.56 = 4.36). The total deposition for reduced N is 3.17 kg N /ha /yr (1.98 × 1.60). The grand total (wet and dry oxidized and reduced) is then about 7.5 kg N /ha /yr.

The model has highly simplified organic N deposition. Note that these values reflect emissions before the NO_x SIP-call, which resulted in substantial reductions in NO_x emissions from point sources over the eastern U.S.

For comparison purposes, a collection of chemical transport models (CTMs) (Dentener et al., 2006) yielded total (wet plus dry) deposition to the whole U.S. of about 3.9 Tg N /yr oxidized Nr and 3.0 Tg N /yr ammoniacal N for current emissions. The fate of NO_x is assumed to be primarily HNO_3 or aerosol NO_3^-; organic N species are generally not modeled in detail. Because this analysis includes Alaska, a better estimate for NO_x for the 48 contiguous states is 4.6 Tg N /yr. The variance among models was about 30% (one standard deviation) for deposition fluxes in regions dominated by anthropogenic emissions. Globally, the calculations from the ensemble of 23 CTMs estimated that 36-51% of all NO_y and NH_x emissions are deposited over the ocean. This load could be important to estuarine N loading estimates, as offshore N is carried inshore by currents or through advective processes.

Deposition estimates from mass balance

From estimated total emissions of Nr compounds and observed or simulated export, a reasonable estimate of rate of deposition can be obtained by mass balance – deposition equals emissions minus export. Although substantial uncertainty (about a factor of two) exists for the emissions of NH_3, NO_x release is reasonably well known. In general, advection in the boundary layer and lofting through convection followed by export at higher altitudes are the two main mechanisms that prevent removal of NO_y and NH_x by deposition to the surface of North America (Luke et al., 1992; Li et al., 2004).

Experimental observations have been conducted over the eastern U.S. for more than two decades (Galloway et al., 1984; Galloway and Whelpdale, 1987; Luke and Dickerson, 1987; Galloway et al., 1988). Most recent estimates (Dickerson et al., 1995; Li et al., 2004; Parrish et al., 2004b; Hudman et al., 2007), agree that annually 7-15% of the emitted NO_x is exported in the lower to mid-troposphere.

CTMs derived small export values – on the order of 30% of the total NO_x emitted into the lower atmosphere

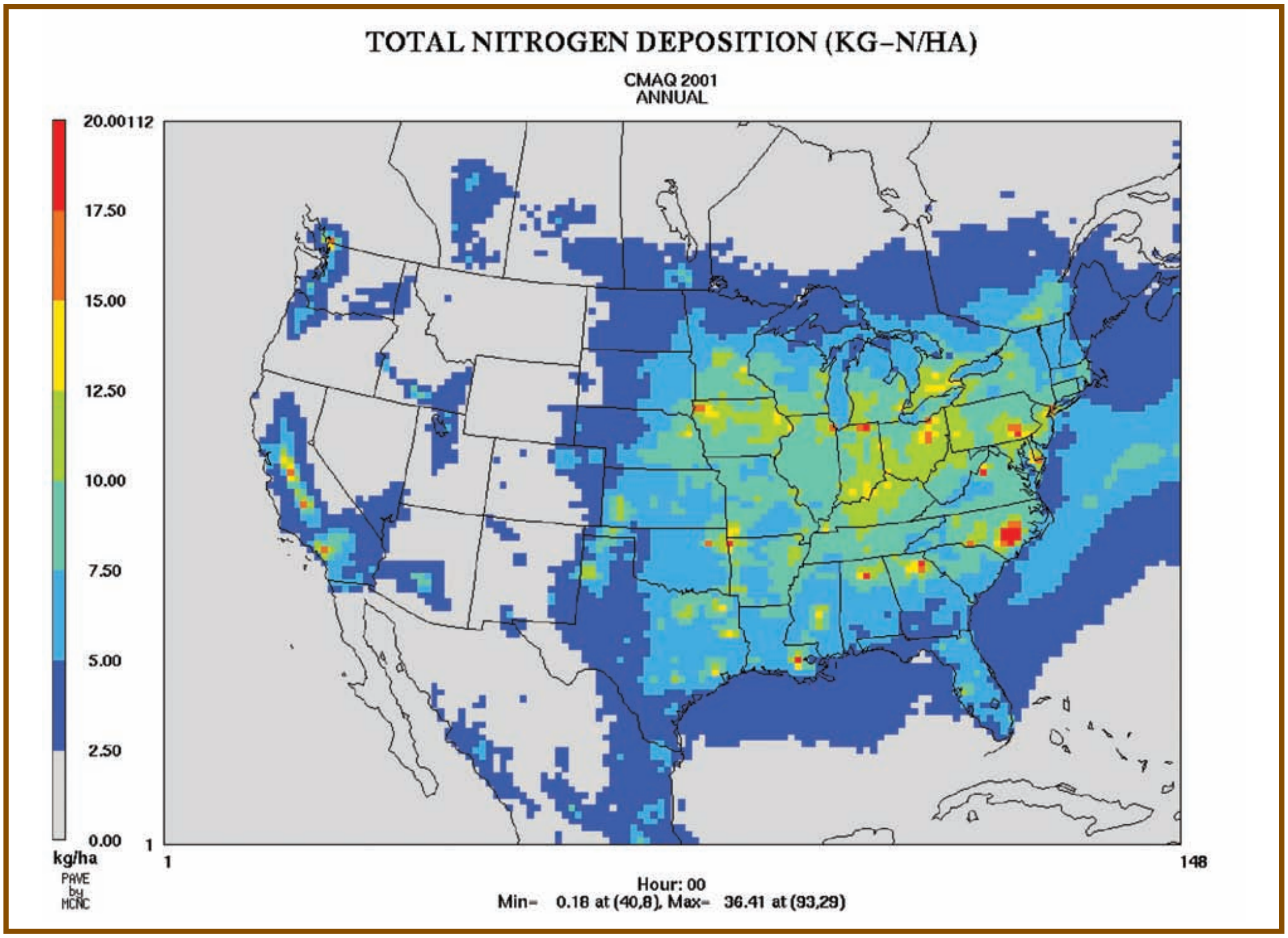

Figure A-4: CMAQ annual average (wet plus dry and oxidized plus reduced) nitrogen deposition (in kg-N/ha/yr) across the United States.

This is based on three years of differing meteorology – one dry, one wet, and one average precipitation year – across the Eastern United States

Source: U.S. Environmental Protection Agency, 2007f

(Penner et al., 1991; Kasibhatla et al., 1993; Holland et al., 1997; Horowitz et al., 1998; Liang et al., 1998; Galloway et al., 2004; Park et al., 2004; Holland et al., 2005; Doney et al., 2007). Reviewed publications using the mass balance approach have substantial uncertainty but indicate with some consistency that 25-35% of the NO_y emitted over the U.S. is exported.

Comparison of models and measurements of oxidized N deposition

Both ambient measurements and numerical models of NO_y have reached a level of development to allow reasonable estimates of deposition. For reduced nitrogen, neither ambient concentrations nor emissions are known well enough to constrain models. Recent model estimates of the U.S. N budget are reasonably uniform in finding that about 25-35% of total NO_x emissions are exported.

Results from CMAQ runs described above indicate that, of the NO_x emitted over the continental U.S., 50% is deposited and 50% is exported. This is within the combined error bars of other studies, but well under the best estimate of 70% deposition. One possible source of this discrepancy is underestimation of deposition of organo-nitrogen compounds. The chemical mechanism used in CMAQ was highly simplified – only about 2-3% of the total Nr deposition can be attributed to organo-nitrogen compounds (R. Dennis personal communication, 2008). Ammonia from fossil fuel combustion, while important locally, is probably a small component of national Nr deposition.

Major sources of uncertainty in modeled and observed values include missing deposition terms and poorly constrained convective mass flux. As indicated above, convective mass flux (rapid vertical transport) is uncertain because most convective clouds are smaller than a grid box in a global model. There is evidence for nonlinearities in NO_2 deposition velocities with greater transfer from the atmosphere to the surface at higher concentrations (Horii et al., 2004; 2006).

Emissions from Canada and Mexico can have a substantive impact on atmospheric Nr over the U.S. near major sources, such as downwind of industrial Ontario and major cities of San Diego, CA, and Tijuana, Mexico (Wang et al., 2009). While Nr is imported into the U.S. from these border countries, there is also export. The emissions from Canada and Mexico are each 10-15% of those of the U.S. and the bulk of the Mexican population is distant from the U.S. We expect the overall impact of neighboring countries to add about 10% uncertainty to the estimated Nr budget for the 48 contiguous states.

Appendix B:
Sources and Cycling of Nr Input into Terrestrial Systems in the United States

Most of the new Nr introduced into terrestrial systems in the U.S. was used to produce food for human consumption and forage and feed for livestock and poultry (~17.7 Tg total with 9.7 Tg from synthetic fertilizer and ~8 Tg from biological N fixation; Table B-1). In addition to new Nr and Nr that was recycled from livestock and human excreta, crop production releases Nr that was stored in soil organic matter (see Section 2.3.2). The N in cereal crops is typically derived from added fertilizer (synthetic or manures) and from mineralization of soil organic matter (conversion of complex organic molecules to ammonium) in about equal amounts. As discussed in Section 2.2 and Section 5.3.4, crop production is not efficient in using Nr so only 30-70% (a global average of 40%) of all the N mobilized for crop production is harvested in the crop. The remainder is in crop residue (roots and above ground stover) stored in the soil, leached to aquatic systems as NO_3^-, volatilized to the atmosphere as NH_3 or NO_x or denitrified (see Section 4.8, Figure 18) to produce NO_x, N_2O and N_2. An additional ~1.1 Tg of synthetic fertilizer N is used to maintain turfgrass in the urban environment (see Section 2.2.5) and another 0.1-0.2 Tg N is used to enhance forest production.

Within the nitrogen cascade (Figure 1), the interactions between the agricultural and populated portions of the terrestrial system dictate the production and flow of Nr. Although occupying the largest area, forest and grassland portions of terrestrial ecosystems serve mainly to absorb atmospheric deposition and provide a source of forest products and forage for livestock production. Reactive nitrogen input into these systems is from biological N fixation in unmanaged lands, atmospheric deposition, and Nr from livestock manure that is deposited. The livestock that is grazing within grasslands (Table B-1) may lead to the N saturation of unmanaged forest and grassland ecosystems (Galloway et al., 2004; Bobbink et al., 2010).

This report uses the Nr input numbers from Table B-1 and food production numbers to estimate the flow of Nr through agricultural and populated parts of the terrestrial system (Table B-2). The FAO (20010b); www.fao.org/es/ess/top/country.html) lists the 20 largest agricultural commodities produced, imported, and exported in the U.S. in 2002. Of these commodities, corn (229 Tg), soybeans (75 Tg), wheat (44 Tg), and cow's milk (77 Tg) were produced in the greatest amount. Using commodity N content data derived from data used to calculate crop

Table B-1: Sources of reactive N into terrestrial systems in the United States in 2002 (from Table 1 data sources; in Tg N/yr).

Source	Environmental System				
	Agricultural	Vegetated		Populated	Total
		Forest	Grassland		
Atmospheric	1.3	1.4	1.9	0.4	6.9*
N fixation	7.7	–	6.4	–	14.1
Synthetic N	9.7	0.1	**	1.1	10.9
Animal manure##	1.2	–	3.8#	–	6.0#
Human sewage##	0.1	–	–	1.2	1.3

*The amount of atmospheric Nr deposition is based on area of each environmental system within the continental U.S. The total area does not sum to 100% because non-arable lands are not included in this table.

**Synthetic fertilizer N used for managed pasture fertilization is included in the agricultural land classification.

#Unrecoverable livestock manure deposited on grasslands, the unaccounted for ~1 Tg of Nr assumed to be lost through ammonia volatilization, leaching, or denitrification (U.S. EPA, 2007e).

##Note that livestock manure and human sewage used as fertilizer are recycled N components of the nitrogen cascade and not new Nr inputs.

residue N in the EPA inventory of U.S. greenhouse gas emissions and sinks (U.S. EPA, 2007e), an estimated 9 Tg of N was marketed in three crops, soybeans (4.4 Tg N; from U.S. EPA, 2007g), corn (3.2 Tg N), and wheat (0.9 Tg N). Whole milk contained ~ 0.5 Tg of N while other meat and egg produce contained ~1.4 Tg of N, totaling ~ 1.9 Tg N. Grain, fruits, nuts and vegetables contained ~9.3 Tg of N. If the total N input use efficiency is 40%, then ~23 Tg of N from all sources is required to produce 9.3 Tg of vegetative commodities. Table B-2 lists the estimated Nr input into agricultural systems (~ 20 Tg) and additional N input from crop residue that was returned to the field the previous year (4.4 Tg) and from mineralization of soil organic matter (4.7 Tg). All of this N input totals ~29 Tg of N that is actually involved in the production of the 9.3 Tg of crop commodity N. If one assumes that return of crop residue to the field is directly proportional to crop production, then 24.3 Tg of N was required to produce the 9.3 Tg of crop commodity N. These estimates indicate that ~38% of the total annual input of N that went into the agricultural crop production system was contained in the main crop commodities produced in the U.S. in 2002.

Of the 24.3 Tg N required to produce crop commodity N in 2002, approximately 2.5 Tg was used to grow feed used for milk, egg, and meat production. This estimate is made assuming that 4 units of N are required to produce a unit of milk, eggs, or meat (see Section 5.3.4). This estimate also assumes that one-third of N required for livestock production comes from commodities in the FAO top-20 list and the remaining two-thirds comes from alfalfa, silage, and grass over the course of a year (Oitjen and Beckett, 1996). Approximately 4.3 Tg of N in agricultural commodities (2.8 Tg in soybeans, corn, and wheat) were exported, while ~0.15 Tg N was imported in various food and drink commodities. The U.S. human populace consumed ~1.96 Tg of N in 2002 (292 million people, consuming 114.7 g protein/person/day, 0.16 g N/g protein, 365 days) (approximately 1.2 Tg from animal protein-N and 0.7 from vegetative protein).

These three consumption areas – internal consumption of vegetable N for livestock production, human consumption, and export – account for 77% of the commodities produced. The unaccounted for commodity N is likely partly in annual storage. Some smaller fraction of annual production is used for pet food and a small fraction is returned to the terrestrial environment because of spoilage and handling losses.

In forests and grasslands (vegetated system) N input in 2002 was ~3.5 Tg of anthropogenically introduced N, with the remaining ~10.1 Tg derived from BNF and livestock manure deposition. Of this anthropogenic N, ~21% was retained in soil and tree biomass, while the remainder was removed in tree harvest (~0.2 Tg, see Section 2.3.2.) or lost to other parts of the environment through NH_3 volatilization and NO_3^- leaching and runoff (Table B-2). Total N input into agricultural systems was ~20 Tg, with ~ 11 Tg being removed as products including the transfer of ~2 Tg N as food to the human population. Almost 40% of the N input into agricultural systems is lost through NH_3 volatilization, nitrification/denitrification, and NO_3^- runoff. The 4.2 Tg of Nr of Haber-Bosch N that is used for industrial feedstock is not included in this assessment. Of the input of ~3.3 Tg of N into the populated system ~80% is lost through human excreta processed in sewage treatment plants, denitrification in soils, and leaching and runoff of NO_3^- (Table B-2).

Table B-2 summarizes the input and flow of Nr in the main terrestrial systems within the continental U.S. Anthropogenic input of Nr into forests and grasslands totaled ~3.5 Tg in 2002, with an estimated 6.4 Tg of Nr being introduced through natural biological N fixation. Of this Nr ~ 0.7 Tg was stored in vegetation and soils (see Section 2.3.2) and ~2 Tg removed as livestock forage, while the remainder was lost to the atmosphere and aquatic systems, or removed as forest products and livestock forage. The largest anthropogenic Nr input (~20 Tg) was into agricultural production where ~11.2 Tg was removed as agricultural product, ~ 2 Tg transferred as

Table B-2: Nr input and flows (Tg N/yr) in the terrestrial portion of the Nitrogen Cascade (Figure 1) within the continental United States in 2002

Environmental System*	N Input to System	N Storage in System**	Agricultural & Forest Products	Transfers to Aquatic or Atmospheric	Transfers as a % of Input
Vegetated	13.6	0.7	2.2	10.7	79
Agricultural	19.6	0.8	11.2	7.6	39
Populated	3.3	0.1	0	3.2	97

*The Environmental Systems are those noted in the terrestrial portion of the N cascade shown in Figure 1. Data from Table B-1, derived from regrouping information from Table 1 data sources, are shown in Table B-2.

**Estimates are from section 2.3.2 of this report.

edible product to the “populated” portion of the terrestrial system, ~0.8 Tg was stored in agricultural lands, and ~7.6 Tg N was lost to the atmosphere and aquatic systems. New N input into the “populated” portion totaled ~3.3 Tg, which came from N transfer in food and use of fertilizer N in lawns, gardens and recreational areas. Within these areas an estimated 0.12 Tg was stored in urban forests.

Appendix C:
Water Quality Trading in the Illinois River Basin

For various reasons, wetland restoration has been proposed and the magnitude of needed restoration estimated. For the Wetland Reserve Program (WRP), the Farm Bill of 1990 set a goal of restoring approximately 1 million acres. A few years later, the NRC (1992) proposed a national goal of restoring 10 million acres of inland and coastal wetlands by 2010. The NRC went on to recommend that 400,000 miles of streams and rivers be restored by 2012 and that 1 million acres of lakes be restored by 2000, both of which would further the control of reactive nitrogen. While none of these goals has been or is likely to be met by the recommended date, they articulated a need for wetland restoration addressing the important relationship between wetlands and water quality.

Taking into account the economics of using wetlands to manage Nr adds yet another dimension to site selection. Based on the results of the Water Environment Research Foundation's study (Hey et al., 2005a,b), the Kinship Foundation sponsored a study (Scott et al., in preparation) to define the market for producing and selling Nr (as NO_3^-) credits. For this analysis, a real potential market area was selected: the Illinois River watershed in Illinois – the tributaries draining Wisconsin, Indiana, and Michigan were excluded. The producers of nitrogen credits were identified as "nutrient farmers" and they became the "sellers" of N credits. The "buyers" of nitrogen credits were restricted to municipal and industrial wastewater treatment facilities, those facilities that hold an NPDES permit. This restriction, of course, resulted in a considerable understatement of the market size since the identified buyers emit less than 11% of the total aquatic N load (David and Gentry, 2000) that finds its way to the Mississippi River – air emission/deposition and agriculture account for the remaining 89%.

The watershed was divided into 19 sub-watersheds, spatially locating credit supply and demand. A linear programming model was developed and used to: (1) examine the potential extent and distribution of nitrogen credit demand and supply; (2) compare the average seasonal demand levels to the supply capacity of nutrient farms; and (3) evaluate the relative effects of seasonality. Market efficiency was imposed through the objective function: the least costly distribution of credit production to meet the given monthly demand. Thereby, sellers and buyers were identified and linked and the spatial characteristics of the market mapped by sub-watershed. At the same time, the equilibrium price of a credit, or the prevailing price at which buyers and sellers are willing to trade, was determined. The market, as represented by the model, determined where the most intensive wetland investment (i.e., wetland restoration) would be, the revenues returned to these investments, and the costs and savings to the buyers.

All 290 permitted buyers are geographically distributed as shown in Figure C-1. The mass loading of the buyers (2,423 tons/month) is reflected in Figure C-2. Eighty-nine percent of the demand comes from the northeastern corner of the Basin (Upper Fox, Des Plaines, and Chicago/Calumet sub-watersheds), the Chicago metropolitan area. As illustrated by Figure C-3, 41% of the wetland restoration area (using the criteria discussed above) was identified in the southwestern corner of the watershed (Lower Illinois, La Moine, Macoupin, Lower Sangamon, and Middle Illinois sub-watersheds), where the floodplain is almost entirely leveed. For the market study, the available load of Nr (NO_3^-) by season and sub-watershed was mapped as illustrated in Figure C-4. The N load was computed using water quality and flow data collected by the U.S. Geological Survey from 1987 to 1997. The wetland and wastewater cost functions are described in Hey et al., 2005; however, the wetland cost functions were modified for the market study to reflect the variability of land costs across the watershed (i.e., higher land values in urban Chicago and lower land values in rural Illinois). This variability is reflected in the spatial distribution marginal costs shown for the spring marginal costs depicted in Figure C-5. Wetland treatment costs vary by time of year because the level of microbial activity, which drives the denitrification process, varies with water temperature. Therefore, treating an equivalent load of Nr requires more wetland area in winter than in summer.

Three regulatory scenarios

Regulatory agencies may require that dischargers and nutrient farms be located in proximity to each other and could impose "penalties" when the two are not. Thus, for the sake of analysis, the Committee created three regulatory scenarios:

1. Unrestricted – buyers can purchase nitrogen credits from nutrient farmers anywhere in the watershed without regard to location. The result of this scenario is given in Figure C-6.
2. Restricted intra-watershed – the buyer must purchase all available credits within its own sub-watershed before buying in other sub-watersheds

3. Accrued 10% penalty – buyers pay an increasing "tax" on credits purchased in consecutive downstream watersheds

The three regulatory scenarios were analyzed for each of the four seasons. (D. Hey, Wetlands Research, Inc., Personal Communication.)

The "unrestricted" scenario is the least expensive because nutrient farms in this scenario are located downstate, where land is least expensive. In the other two scenarios, credits were purchased a little more evenly throughout the watershed. Still, most of the credits in the southern corner of the watershed were purchased. The

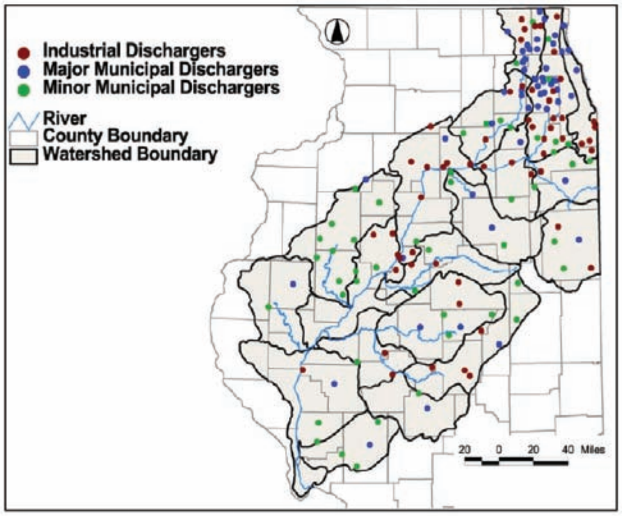

Figure C-1: Distribution of municipal discharge and industrial dischargers in the Illinois River Watershed

Municipal dischargers shown are those exceeding one million gallons per day. Symbols may represent more than one discharger at that location.

Source: D. Hey, Wetlands Research, Inc., Personal Communication

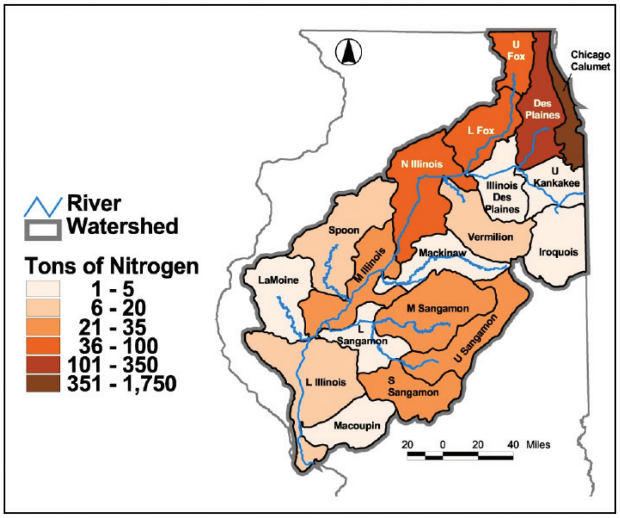

Figure C-2: Distribution of total nitrogen emissions by sub-watershed

Source: D. Hey, Wetlands Research, Inc., Personal Communication

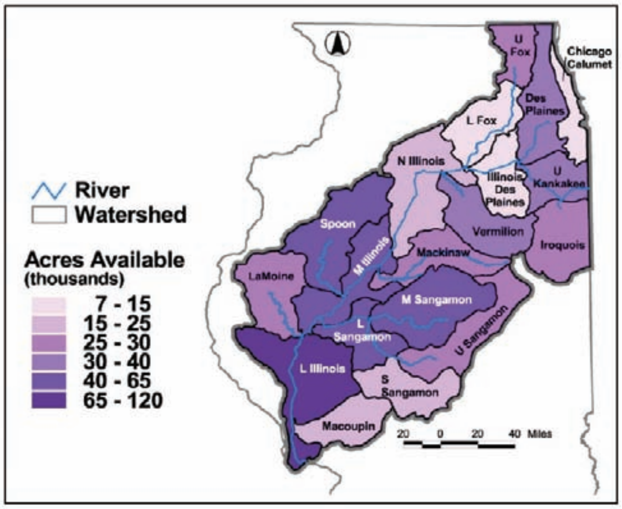

Figure C-3: Potential land availability in the 100-year flood zone for nutrient farming in each sub-watershed in the Illinois River watershed

Source: D. Hey, Wetlands Research, Inc., Personal Communication

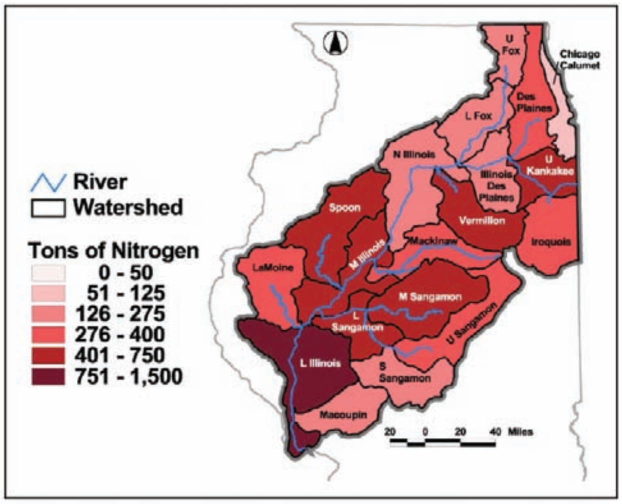

Figure C-4: Spring available total nitrogen load by sub-watershed

Source: D. Hey, Wetlands Research, Inc., Personal Communication

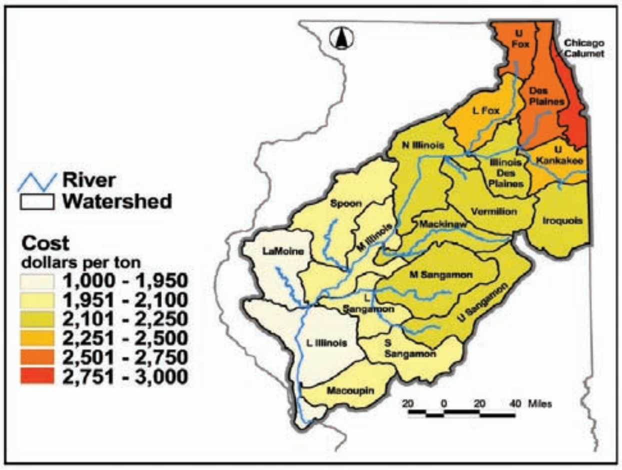

Figure C-5: Spring marginal cost (price) by watershed

Source: D. Hey, Wetlands Research, Inc., Personal Communication

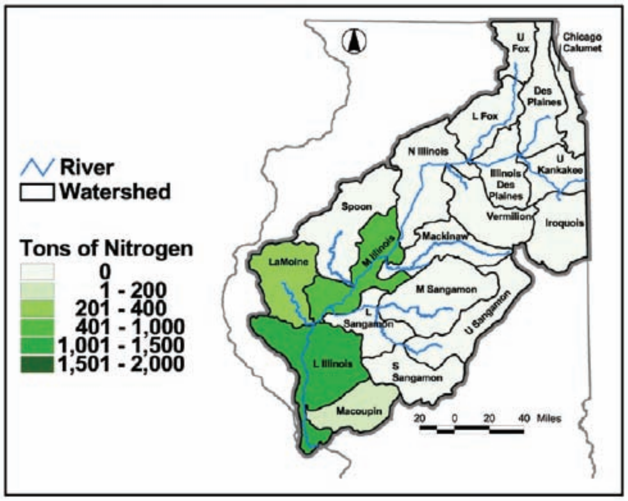

Figure C-6: Unrestricted spring credit sales (tons/month) by sub-watershed

Source: D. Hey, Wetlands Research, Inc., Personal Communication

"restricted intra-watershed" and "accrued 10% penalty" scenarios resulted in more credits being purchased. This resulted in the sale of N credits exceeding the mass of Nr emitted by wastewater treatment, which would benefit the overall control of reactive nitrogen. It also would increase the value of the market and the profits of the nutrient farmer. The downside of such regulatory controls is that they would drive up the effective price of nitrogen credits. If a buyer had to buy a 1.5 tons for every ton discharged because credits are not available in the tributary watershed, the effective price of a credit would be 1.5 times the price of the tributary sub-watershed. If prices rise too much, "concrete and steel" technologies may become competitive.

Considering all of the point source dischargers in the Illinois River watershed, between 29,000 and 36,000 tons TN/year could be removed through nutrient farming under the studied trading schemes (Table C-1). The range of removal is a function of the penalties imposed on the market by the regulatory agencies. Accordingly, the market revenue would range from $70 million to $121 million/year. This is a sizeable market that could generate substantial profits, from $6 million to $38 million with the return on investment varying from 5 to 25%. If the savings are shared evenly between the seller and buyer, the nutrient farmer could earn between $200 and $300/acre/year net profit, which in many cases is greater than the profits from corn or soybean production. Further, these profits do not include any earnings from flood control or recreation, as suggested in a McKnight study report (Hey et al., 2004). With such profits, sufficient land should be available for nutrient farming.

This analysis indicates that appropriate lands are available and that wetlands can be effectively restored and efficiently used to control Nr. The market, structured as discussed above, could generate the capital to accomplish the needed large-scale wetland restoration while saving taxpayers the cost of upgrading their municipal wastewater treatment plant (TWI, 2007).

Table C-1: Nutrient farm market parameters under three trading scenarios

Parameter	Unrestricted	Restricted Intra-watershed	Accrued 10% Penalty
Total Credits Sold (tons)	29,078	29,078	35,781
Total Revenue [21]	$69,925,497	$99,571,889	$121,457,652
Total Cost to Produce Credits	$63,258,006	$66,193,924	$83,288,747
Profit	$6,667,491	$33,377,968	$38,168,905

Source: D. Hey, Wetlands Research, Inc., Personal Communication

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[21] Assumes all credits were sold at the cheapest cost within the Illinois River watershed.

Appendix D:
Management of Nr Measures Based on the Concept of Critical Loads

The European Union has undertaken broad measures, based on the critical-loads concept, to manage Nr. Tables D-1, D-2, and D-3 summarize several different environmental impacts currently used as indicators and identify whether there are current limit values set by the United Nations Economic Commission for Europe (UNECE) or European Union (EU). These tables identify the main links to the cascade of reactive nitrogen in the environment, the relevance and link to Nr of the effect/pollutant, and existing agreements in which the effect is currently addressed. In addition, some impacts are more relevant than others in relation to societal importance and the connection to the nitrogen cascade. The categorization on a scale of 1 (highest relevance) to 5 (unimportant) provides a first-level prioritization for future mitigation activity. The last column summarizes existing links to international regulations and conventions.

Where there is a limit and the relevance for the nitrogen cascade is high, then this might be the limiting factor for Nr production and its associated losses to the environment. Some limits might be more relevant in specific areas and less relevant in others. For example NO_2 concentrations relevant for human health are limited to 40 ppb in urban areas, limiting industry and traffic, but would probably not be an issue of concern in remote areas with low population densities. In these areas, however, loss of biodiversity might limit nitrogen deposition and therewith the sources in the region. The only way to determine the extent that critical thresholds are limiting is by overlaying them on different regions and determining, through the use of monitoring data or by modeling exercises, where and which sources contribute to exceeding the critical threshold. Then the best methods for putting caps on relevant sources can be identified. A pre-classification of regions might be useful, e.g., urban regions, remote regions, marine areas, etc. One aspect of this global view of nitrogen impacts and metrics that is evident is the mix of “classical-” and “service”-based categories, consistent with the need for an integrated approach to the management of nitrogen.

Table D-1: Summary of the effects of excess Nr on human health in relation to metrics, current international regulations and conventions, and the link to the nitrogen cascade

	Metrics	Regulated?	Link to Nr cascade	Relevance*	Regulatory or political convention
Respiratory disease in people caused by exposure to high concentrations of:					
Ozone	Sum of ozone over 35 ppb	YES	NO_x emissions	3	Convention on Long-range Transboundary Air Pollution Clean Air for Europe
Other photochemical oxidants	Org. NO_3, PAN concentration (atm)	NO	NO_x emissions	5	Convention on Long-range Transboundary Air Pollution et al.
Fine particulate aerosol	PM_{10}, $PM_{2.5}$ concentration (atm)	YES	NO_x, NH_3 em	1	Convention on Long-range Transboundary Air Pollution Clean Air for Europe
Direct toxicity of nitrite NO_2^-	NO_2^- concentration	YES	NO_x	2	World Health Organization Convention on Long-range Transboundary Air Pollution Clean Air for Europe
Nitrate contamination of drinking water	NO_3^- concentration (aq.)	YES	NO_3^- leaching	2	EU Essential Facilities Doctrine
Depletion of stratospheric ozone	NO_x, N_2O concentration/ flux (atm)	NO	NO_x, N_2O	3	Montreal Protocol
Increase allergenic pollen production, and several parasitic and infectious human		NO		5	None
Blooms of toxic algae and decreased swimability of in-shore water bodies	Chlorophyll a NO_3^- (&P) concentration (aq)	NO	Runoff, Nr deposition	1	Convention for the Protection of the Marine Environment of the North-East Atlantic Helsinki Commission Barcelona Convention

atm – atmospheric; aq – aqueous

*Relevance and link to nitrogen incorporates societal priority and N contribution: 1. highest relevance, 2. high relevance, 3. significant relevance, 4. some relevance, 5. unimportant.

Table D-2: Summary of the effects of excess Nr on ecosystems related to currently used metrics, the existence of European regulatory values, and the link to the nitrogen cascade.

	Metrics	Regulated?	Link to Nr cascade	Relevance*	Regulatory or political convention
Ozone damage to crops, forests, and natural ecosystems	AFstY (O_3 flux), AOT40**	YES	NO_x	2	Convention on Long-range Transboundary Air Pollution Clean Air for Europe
Acidification effects on terrestrial ecosystems, ground waters, and aquatic ecosystems	Critical loads	YES	Nr deposition	2	Convention on Long-range Transboundary Air Pollution Clean Air for Europe WFD
Eutrophication of freshwaters, lakes (incl. biodiversity)	Biological Oxygen Demand, NO_3^- conc (aq) Critical loads	YES NO	Runoff, Nr deposition	3	Water Framework Directive
Eutrophication of coastal ecosystems inducing hypoxia (incl. biodiversity)	BOD, NO_3^- conc (aq) Critical loads	BOD, NO_3^- conc (aq) Critical load	Runoff, Nr deposition	1	Convention for the Protection of the Marine Environment of the North-East Atlantic Helsinki Commission Barcelona Convention
Nitrogen saturation of soils (incl. effects on GHG balance)	Critical loads	YES	Nr deposition	1	Convention on Long-range Transboundary Air Pollution Clean Air for Europe
Biodiversity impacts on terrestrial ecosystems (incl. pests and diseases)	Critical loads, critical levels (NH_3, NO_x)	YES	Nr deposition	1	Convention on Long-range Transboundary Air Pollution Clean Air for Europe Convention on Biological Diversity

atm – atmospheric; aq – aqueous

*Relevance and link to nitrogen incorporates societal priority and N contribution: 1. highest relevance, 2. high relevance, 3. significant relevance, 4. some relevance, 5. unimportant.

**Accumulated ozone exposure over a threshold of 40 parts per billion

Table D-3: Summary of the effects of excess N on other societal values in relation to metrics and regulatory values in current international regulations and conventions and the link to the nitrogen cascade

	Metrics	Regulated?	Link to Nr cascade	Relevance*	Regulatory or political convention
Odor problems associated with animal agriculture	Acidity in precipitation., O_3, PM	YES	NO_x, NH_3	3	Convention on Long-range Transboundary Air Pollution
Effects on monuments and engineering materials	$PM_{2.5}$ conc (atm)	NO	NO_x, NH_3	4	
Global climate warming induced by excess nitrogen	N_2O, conc/flux (atm)	NO	NO_x, NH_3	1	United Nations Framework Convention on Climate Change
Regional climate cooling induced by aerosol)	$PM_{2.5}$ conc (atm)	NO	NO_x, NH_3	1	United Nations Framework Convention on Climate Change

atm – atmospheric; aq – aqueous

*Relevance and link to nitrogen incorporates societal priority and N contribution: 1. highest relevance, 2. high relevance, 3. significant relevance, 4. some relevance, 5. unimportant.

Appendix E:
Technical Annexes

Production of N_2 and N_2O via gas-phase reactions

Atmospheric conversion of NO_x and NH_x to less reactive N_2 or N_2O appears to play a minor role in the global N budget, but currently is not well quantified. The gas-phase reactions in the troposphere that convert NH_3 and NO_x to N_2 and N_2O, start with attack of NH_3 by OH:

$$NH_3 + OH\cdot \rightarrow NH_2\cdot + H_2O \quad (1)$$

Several potentially interesting fates await the NH_2 radical:

$$NH_2\cdot + O_3 \rightarrow NH, NHO, NO \quad (2)$$

$$NH_2\cdot + NO_2 \rightarrow N_2O + H_2O \quad (3)$$

$$NH_2\cdot + NO \rightarrow N_2 + H_2O \quad (4)$$

$$kO_3 = 1.9\text{x}10^{-13}\ cm^3\ s^{-1}$$

$$kNO_2 = 1.8\text{x}10^{-11}\ cm^3\ s^{-1}$$

$$kNO = 1.8\text{x}10^{-11}\ cm^3\ s^{-1}$$

The first step, attack by OH, is slow. The rate constant for the Reaction 1 is 1.6 x 10^{-13} cm^3 s^{-1} and the lifetime of NH_3 for a typical concentration of 106 OH cm^{-3} is about 70 days. In most areas of the world where concentrations of NH_3 are high, concentrations of sulfates are also high, and NH_3 is removed by conversion to condensed phase ammonium sulfate or bisulfate on time scales much faster than 70 d. The mean lifetime of these aerosols with respect to wet deposition is about 10 d.

There are some areas of the world, notably California and South Asia, where NH_3 and NO_x are emitted in large quantities, but SO_2 is not, and there gas-phase conversion can take place. NH_3 is usually removed by wet or dry deposition. Reaction 2 is relatively slow and oxidation of ammonia is in general an unimportant source of NO_x. But Reactions 3 and 4 may be atmospherically noteworthy. As an upper limit to current N_2O production, we can assume that each of these regions covers an area of 10^6 km^2 and that they contain ammonia at a concentration of 10 g N m^{-3} in a layer 1,000 m deep. The annual production of N_2 and/or N_2O would then be on the order of 0.1 Tg N, a minor but nontrivial contribution to denitrification and about 1% of the anthropogenic N_2O production. If NH_3 rich air is lofted out of the boundary layer into the upper troposphere where deposition is impeded, it will have an atmospheric residence time on the order of months, and the probability of reaction to form N_2O or N_2 becomes greater. This possibility has not been investigated extensively. It is also possible than Europe and North America will continue to reduce S emissions without reducing NH_3 emissions and the atmospheric source of N_2O will grow in importance.

In the stratosphere, N_2O photolysis leads to loss of Nr via

$$N_2O + h\mu \rightarrow N_2 + O \quad (5)$$

While reaction with an electronically excited oxygen atom O(1D) leads to production of NO via

$$N_2O + O(^1D) \rightarrow 2NO \quad (6)$$

Photolysis (Reaction 5) dominates, but a large enough fraction of the N_2O reacts with $O(^1D)$ that this is the main source of NO_x in the stratosphere. The fate of this oxidized nitrogen (NO_y) is transport back into the troposphere where it is removed by wet deposition. Downward transport of the odd N from the oxidation of N_2O is a minor (~1%) source of NO_y in the troposphere. Most of the N_2O released into the atmosphere is eventually converted to N_2 – the problem is that it destroys stratospheric ozone in the process.

In summary, our current understanding of the chemistry of atmospheric ammonia suggests that *in situ* conversion to N_2 and N_2O plays a minor (~1%) role in global N budgets, but if assumptions about kinetics or concentrations are in error, these mechanisms could become important.

SPARROW model for estimating watershed Nr

Estimates of Nr transfers in aquatic ecosystems are difficult to quantify at the national scale, given the need to extrapolate information from sparse monitoring data in specific watersheds to the geographic boundaries of the nation. One excellent tool for estimating Nr loads at regional scales is the spatially referenced regression on watershed attributes (SPARROW) modeling technique. The SPARROW model has been employed to quantify nutrient delivery from point and diffuse sources to streams, lakes, and watershed outlets at the national scale (Smith et al., 1997). The model infrastructure operates in a geographic framework, making use of spatial data to describe sources of pollutants (e.g., atmospheric deposition, croplands, fertilizers) and characteristics of the landscape that affect pollutant transport (e.g., climate, topography, vegetation, soils, geology, and water routing). Though empirical in nature, the SPARROW modeling approach uses mechanistic formulations (e.g., surface-water flow paths, first-order loss functions), imposes mass balance constraints, and provides a formal parameter estimation structure to statistically estimate sources and fate of nutrients in terrestrial and aquatic ecosystems. The spatial referencing of stream monitoring stations, nutrient sources, and the climatic and hydrogeologic properties of watersheds to stream networks explicitly separates landscape and surface-water features in the model. This allows nutrient supply and attenuation to be tracked during water transport through streams and reservoirs, and accounts for nonlinear interactions between nutrient sources and watershed properties during transport. The model structure and supporting equations are described in detail elsewhere (Smith et al., 1997, Alexander et al., 2000, Alexander et al., 2008). Figure E-1 provides an estimate of contemporary Nr loading in surface waters of the U.S., representing long-term average hydrological conditions (over the past three decades). There are hot spots of high Nr yields to rivers associated with land use and watershed characteristics, and SPARROW allows considerations of the fate of these Nr inputs to streams and rivers as they flow downstream to coastal receiving waters (Alexander et al., 2008).

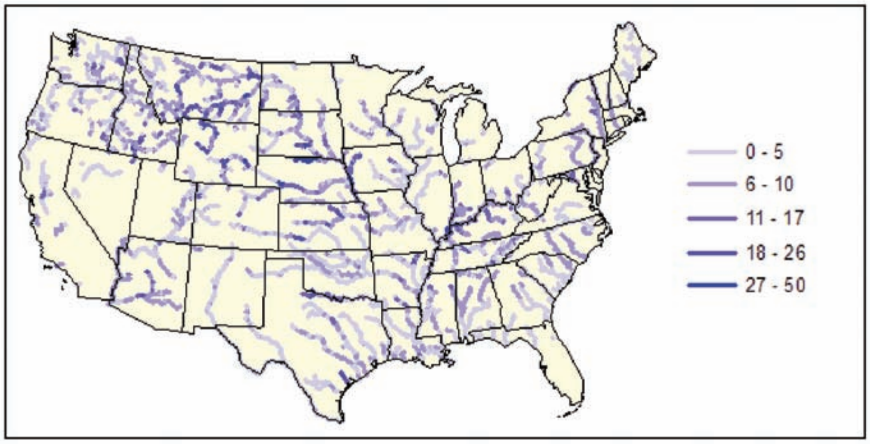

Figure E-1: Total Nr yields (kg/ha/yr) in large rivers of the U.S.

Data Source: Alexander et al., 2008

Appendix F:
Recent Major EPA Mobile Source Rules to Control NO_x

EPA informed the Committee that it is in the process of implementing a number of regulations to reduce NO_x from a variety of mobile sources[22]. These include clean diesel regulations for trucks and buses and nonroad engines, as well as locomotives and smaller marine vessels. EPA first regulated NO_x emissions from motor vehicles for the 1973 model year and since then has tightened these standards. EPA's efforts to control NO_x emissions from nonroad vehicles, locomotives, and commercial marine vessels started in the 1990s. NO_x reductions for each rule were calculated by EPA based on inventories available at the times of the rules.

1. Light Duty Tier 2 Rule – EPA's Tier 2 Vehicle and Gasoline Sulfur Program (65 FR 6698, February 10, 2000). This program requires new cars, sport utility vehicles (SUVs), pickup trucks, and vans to be 77 to 97% cleaner than 2003 models, while reducing sulfur levels in gasoline by 90%. EPA estimates that as newer, cleaner cars enter the national fleet, the new tailpipe standards will reduce emissions of nitrogen oxides from vehicles by 3 million tons, or about 74% in 2030. Prior to that, the EPA Tier 1 vehicle regulations, effective with the 1995 model year, also resulted in significant NO_x reductions.
2. EPA's Clean Heavy Duty Truck and Bus Rule. When the Agency finalized the Heavy Duty Truck and Bus Diesel Rule (66 FR 5002, January 18, 2001) in 2001, trucks and buses accounted for about one-third of NO_x emissions from mobile sources. In some urban areas, the contribution was even greater. With model year 2010, all new heavy duty trucks and buses will result in NO_x emission levels that are 95% below the pre-rule levels. EPA projects a 2.6 million ton reduction of NO_x emissions in 2030 when the current heavy-duty vehicle fleet is completely replaced with newer heavy-duty vehicles that comply with these emission standards.
3. Clean Air Nonroad Diesel – Tier 4 Rule (69 FR 38957, June 29, 2004). In 2004, EPA adopted a comprehensive national program to reduce emissions from future nonroad diesel engines by integrating engine and fuel controls as a system to gain the greatest emission reductions. EPA estimates that in 2030, this program will reduce annual emissions of NO_x by about 740,000 tons.
4. Marine-Related NO_x Reductions from 1999 to 2003. EPA completed three rulemakings with respect to the diesel marine sector that will reduce NO_x emissions. These rules are now in effect and being phased-in. In 1999 (64 FR 73299, December 29, 1999), EPA promulgated NO_x requirements for diesel engines used in commercial boats (large inland and near-shore boats) and commercial vessels (ocean-going vessels). EPA estimates that these reduced emissions from these vessels by about 30%. In 2002 (67 FR 68241, November 8, 2002), EPA promulgated rules reducing NO_x emissions from diesel engines used in recreational marine vessels by 25%. In 2003 (68 FR 9746, February 28, 2003), EPA promulgated another rule further reducing NO_x from diesel engines used in commercial vessels by about 20%. EPA projects that on a nationwide basis, these four programs will reduce marine-related NO_x by more than 1 million tons in 2030.
5. Locomotive and Marine Diesel Rule (73 FR 25098, May 6, 2008). In March 2008, EPA adopted standards that will reduce NO_x emissions from locomotives and marine diesel engines. The near-term emission standards for newly-built engines phased in starting in 2009. The long-term standards begin to take effect in 2015 for locomotives and in 2014 for marine diesel engines. EPA estimates NO_x emissions reductions of 80% from engines meeting these standards. EPA projects that in 2030, about 420,000 tons of NO_x will be reduced from the locomotive engines, and 375,000 tons of NO_x will be reduced from commercial and recreational marine engines.
6. Non-road Spark-Ignition Engines (73 FR 59034, October 8, 2008). In 2002, EPA promulgated emissions standards for large spark-ignition engines. These took effect in 2004 for Tier 1 standards and in 2007 for Tier 2 standards. EPA promulgated emissions standards for small spark-ignition engines in 2008. EPA projects that, when fully implemented, the new standards will result in a 35% reduction in $HC+NO_x$ emissions from new engines' exhaust, reduce evaporative emissions by 45%, and that together these programs will reduce NO_x by more than 585,000 tons in 2030.
7. EPA's Coordinated Strategy for Control of Emissions from Ocean-Going Vessels (www.epa.gov/otaq/oceanvessels.htm). EPA's coordinated strategy to

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[22] The information in Appendix F was provided to the Integrated Nitrogen Committee by Mazrgaret Zawacki of the U.S. EPA Office of Transportation and Air Quality.

control emissions from ocean-going vessels consists of actions at the national and international levels. On December 22, 2009, EPA finalized emissions standards for ocean-going vessels which take effect in 2011. In addition to this rule the U.S. Government has also amended MARPOL Annex VI to designate U.S. coasts as an Emission Control Area (ECA) in which all vessels, regardless of flag, will be required to meet the most stringent engine and marine fuel sulfur requirements in Annex VI. New engine emission and fuel sulfur limits contained in the amendments to Annex VI are also applicable to all vessels regardless of flag and are implemented in the U.S. through the Act to Prevent Pollution from Ships (APPS). EPA projects that when fully implemented, the coordinated strategy will reduce NO_x emissions from ocean-going vessels by 80% and that in 2030, the coordinated strategy is expected to yield a reduction in NO_x of about 1.2 million tons.

8. EPA's Voluntary Clean Diesel Programs. EPA has created a number of programs designed to reduce emissions (including both PM and NO_x) from the diesel fleet. In conjunction with state and local governments, public interest groups, and industry partners, EPA has established a goal of reducing emissions from the over 11 million diesel engines in the existing fleet by 2014. Looking at these engines, EPA determined there were general sectors that provided the best opportunity to obtain significant reductions and created programs for Clean Agriculture, Clean Construction, Clean Ports, Clean School Bus, and SmartWay Transport.
9. Section 177 of the Clean Air Act allows states outside of California to adopt California emissions standards, once EPA has granted such a waiver. As a result, several northeastern states have adopted California standards. Maryland adopted its California LEV II NO_x standards as part of its Low Emission Vehicle Program (COMAR 26.11.34, effective December 17, 2007). These standards take effect with the 2011 model year. Maryland submitted that program to EPA as a SIP revision. Pennsylvania adopted California LEV II NO_x standards as part of its Clean Vehicles Program (codified at Pa. Code Chapters 121 and 126, effective December 9, 2006). Pennsylvania's program began with model year 2008 vehicles. Pennsylvania submitted this program as a SIP revision.

Appendix G:
Impacts of Reactive Nitrogen on Aquatic Systems

Nitrogen contamination of groundwater

In addition to environmental concerns about N-nutrient loading to freshwaters from a groundwater pathway, there are also potential human health impacts from elevated levels of N in groundwater, especially from NO_3. It has been long established that excess NO_3 in drinking water supplies can cause blue baby syndrome (methemoglobinemia) (Knobeloch and Proctor 2001; Ward et al. 2005, 2006), the indicator of which is MetHb. To protect public health from effects of NO_3 in drinking water, EPA has established a maximum contaminant level goal (MCLG) that considers a lifetime exposure plus a margin of safety. For NO_3 in drinking water, the MCLG is 10 mg/L. Nitrite-N also has an established MCLG of 1 mg/L, and the combined NO_3 and NO_2 MCLG set by EPA is 10 mg/L. These same values are used to regulate NO_3 and NO_2 as maximum contaminant levels (MCL), which are the highest levels of contaminants allowed in drinking water (40 CFR § 141.62).

The drinking water standard is commonly exceeded in streams and rivers of the U.S., particularly in the agricultural Midwestern U.S. For example, there were 13 episodes over a 25- year period of formal warnings by authorities to local citizens in Columbus, Ohio, about not drinking tap water because nitrate-nitrogen was higher than 10 mg-N/L (Mitsch et al., 2008). These episodes lasted from one to several weeks each. The pattern is generally for high concentrations of nitrate-nitrogen in Midwestern rivers from February through June or July. In one pattern, averaged over 7 years with weekly river sampling, nitrate-nitrogen in a central Ohio river peaked with an average of 7 mg-N/L in June after which concentrations decrease to 1-2 mg-N/L for the rest of the summer and fall. The year-to-year variability was high for that month as well (Mitsch et al. 2005) as these spring "high-nitrate" floods do not occur every year. The nitrate "pulses" generally are part of flood events after fertilizer has been applied to fields in the watershed.

Public policy by water supply agencies is to treat high concentrations of nitrate-nitrogen in drinking water supplies as a real public health threat. Recent studies have brought the concern of high nitrate-nitrogen in drinking water into dispute (Ward et al., 2005, 2006). While there is a definite link between excessive nitrate in drinking water and methemoglobinemia, there is also a need to better understand the interaction of the range of environmental factors (e.g., cofactors such as diarrhea and respiratory diseases reportedly increase MetHb levels) that promote methemoglobinemia. This will help identify the environmental conditions under which exposure to nitrate in drinking water poses a risk of methemoglobinemia.

According to the USGS (Barber, 2009) *Summary of Estimated Water Use in the U.S. in 2005,* total water withdrawals in the U.S., excluding thermoelectric power usage, were 210 billion gallons per day, of which 44,200 million gallons per day (MGD) were for public water supply. About two-thirds of that supply is provided by surface water, the rest is from wells and about 58% (25,600 MGD) of public water supply goes towards domestic use, including drinking water. Private wells (Figure G-1) that are not part of public water supply systems are estimated to provide an additional 3,830 MGD, providing domestic water for 42.9 million people (14% of the U.S. population in 2005).

Groundwater N in forested and low intensity (<10%) agriculture or urban land use areas is estimated to be fairly low, having 75th percentile concentrations of 0.5 and 1.1 mg/L in two USGS studies (Nolan and Hitt, 2002). They consequently concluded that a "reasonable" background concentration, as NO_3-N, would be 1.1 mg/L, which would include effects in more sensitive aquifers with nominal loading for urban or agricultural sources.

Nitrate can enter groundwater from a variety of sources, including all of those described in this report, but fertilizer and animal waste in rural, agricultural areas are especially prominent sources (Nolan and Ruddy, 1996). Other sources include septic systems, more important in densely-developed and unsewered urban areas, and atmospheric deposition. Vulnerability to elevated NO_3 levels is also variable, but an assessment and model by Nolan and Hitt (2006) predicted that "…areas with high N application, high water input, well-drained soils, fractured rocks or those with high effective porosity, and lack of attenuation processes…" are especially vulnerable ($r^2 = 0.801$).

As illustrated in Figure G-2, surveys confirm that NO_3 in groundwater is elevated in many areas of the U.S., well above the 1.1 mg/L background upper bound described above (Figure G-2). The background colors on the map in Figure G-2 are indicative of different types of aquifers. In a 1992-1995 survey (Nolan and Stoner, 2000), shallow groundwater underlying agricultural areas was found to be most severely impacted by elevated NO_3-N levels (median concentration of 3.4 mg/L). Urban shallow aquifers were less impacted (median concentration of 1.6 mg NO_3-N/L) and deeper, major aquifers, in general, had a median NO_3-N concentration of 0.48 mg/L. However, NO_3-N concentrations did exceed the 10 mg/L MCL threshold set by EPA for drinking water in more than 15% of the groundwater samples in the survey from drinking water aquifers.

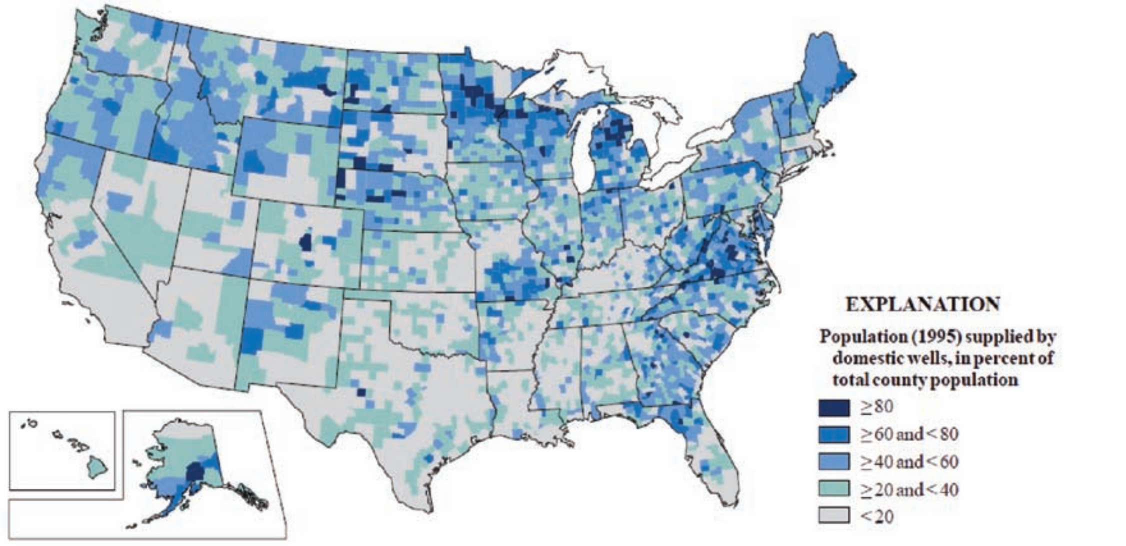
In the most recent survey of domestic well water quality (DeSimone et al., 2009), USGS found concentrations of NO_3-N greater than 10 mg/L in 4.4% of the wells sampled. Concentrations exceeding the nitrate MCL were most frequently encountered in certain basins of the Southwest and California, west-central glacial aquifers in the Upper Midwest, and coastal plain aquifers and Piedmont crystalline rock aquifers in central Appalachia. Lowest concentrations were found in the coastal plain aquifers of the Southeast. In general, higher NO_3-N concentrations were found near agricultural lands. In an additional analysis of shallow groundwater wells in agricultural areas, separate from the national survey, nearly 25% of the sampled wells exceeded the 10 mg/L MCl for NO_3-N. DeSimone et al. (2009) suggested that redox could be a defining factor in some cases, and perhaps was the reason for low NO_3-N concentrations in the Southeast in soils that promote denitrification, as well as higher NO_3-N levels in other areas where aquifers were better oxygenated.

Ammonia toxicity in freshwater systems

The EPA and states have long regulated ammonia (NH_3) in the environment, not because of its nutrient contribution to cultural eutrophication, but because of its toxicity to freshwater aquatic life (U.S. EPA, 1986). The un-ionized ammonia molecule has been identified as the primary toxic form, rather than the ammonium ion (NH_4^+), and research has further demonstrated the relationship between pH, temperature, and NH_3 partitioning from the total ammonia pool in freshwaters (U.S. EPA, 1999). Because of the relationship to temperature in particular, and the variable sensitivity of species and life stages of aquatic organisms, state water quality standards in application generally consider cold and warm water conditions, as well as acute and chronic exposures to life stages of sensitive organisms, plus a margin of safety to derive criteria.

While water quality criteria were initially set for concentrations of NH_3, as criteria development skills and understanding improved, it made sense to develop criteria for total ammonia concentration for specific water quality conditions, e.g., cold or warm (salmonids present or absent) with consideration of ambient pH factors as appropriate, for protection of the most sensitive species likely to be present (early life stages present or absent), plus a margin of safety. This was because of evidence that the NH_4^+ fraction may also be contributing to toxicity. Criteria could be presented as formulas in adopted state criteria to calculate total ammonia thresholds based on prevailing pH and temperature conditions and organisms present/absent, as appropriate.

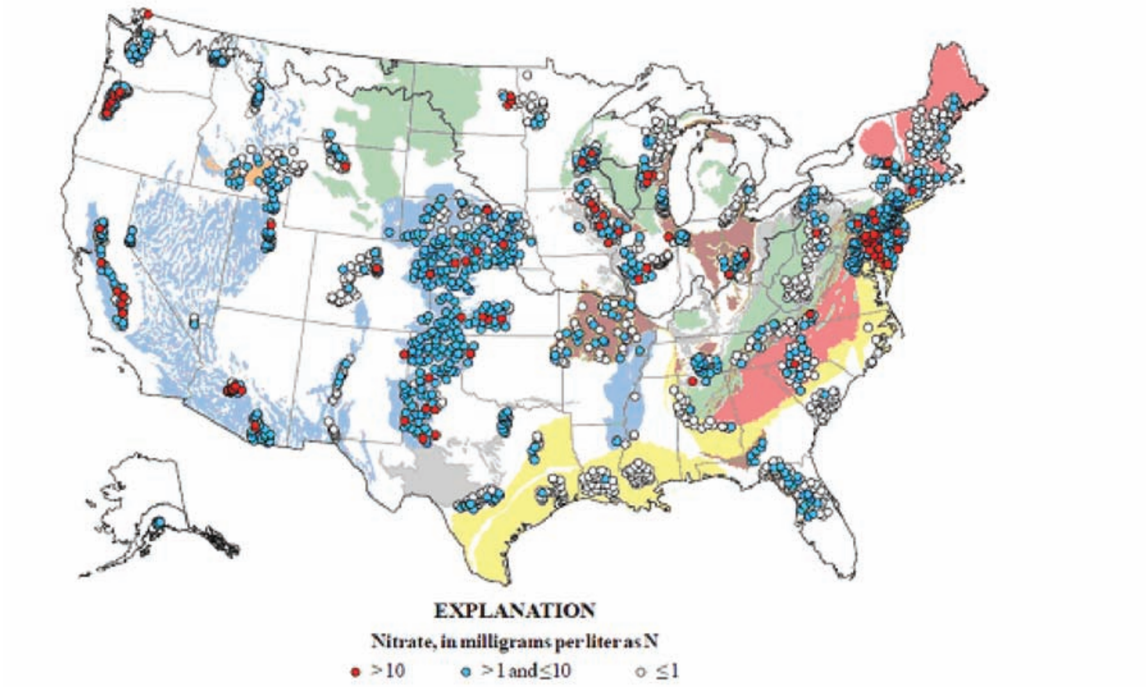
Based on this research and analysis, EPA currently recommends adoption of ammonia criteria as criterion continuous concentration (CCC) and criterion maximum concentration (CMC) as described in Box G-1 (U.S. EPA, 1999).



In many parts of the United States, domestic wells supply drinking water for large percentages of the population. Nationwide, more than 43 million people rely on domestic wells. Data shown are from Solley and others (1998).

Figure G-1: U.S. Population (1995) supplied by domestic drinking water wells

Source: DeSimone et al., 2009.



Concentrations of nitrate were greater than the U.S. Environmental Protection Agency Maximum Contaminant Level of 10 milligrams per liter (mg/L) as N in 4.4 percent of the wells. Elevated concentrations of nitrate occurred throughout the United States, but least commonly in the Southeast. Concentrations greater than 1 mg/L as N, which is usually indicative of human activities in many areas (Nolan and Hitt, 2003), were found in about 40 percent of wells located throughout the sampled areas.

Figure G-2: Nitrate concentrations in U.S. domestic drinking water wells

(Background colors on the map are indicative of different aquifer types)
Source: DeSimone et al., 2009

Recent research on the sensitivity of freshwater unionid mussels (Family, Unionidae), and the rare and endangered status of several unionid species, has led the EPA to issue a draft update to the ammonia criteria guidance (U.S. EPA, 2009b) that would supersede the current 1999 guidance. Mussels have been found to be more sensitive to ammonia toxicity than the most sensitive species used to derive the 1999 criteria (Augspurger et al., 2003). Augspurger et al. (2003) found that the CMC with unionids considered would range from 1.75 to 2.50 mg total ammonia-N/L, 60% lower than the current method calculation of 5.62 mg total ammonia-N/L, for example. In the draft 2009 update, EPA proposed a two-tiered process for ammonia CMC and CCC development for waters with and without sensitive unionid species.

With this new sensitivity identified, ecosystem imbalances due to eutrophication and the presence and die off of invasive species may result in toxic levels of ammonia for mussels in freshwater systems, in addition to the conventional sewage and agricultural sources. For example, Cooper et al., (2005) suggested that die offs of invasive Asian clam (Corbicula fluminea), common in most southeastern U.S. waters, could produce enough sediment pore water ammonia during decay to be lethal to sensitive unionid mussels. Further, combination of ammonia with other toxic substances may compound toxic effects, as found by Wang et al. (2007a,b) for ammonia and copper. Their research suggests that the 1999 criteria for total ammonia might not be protective of sensitive mussel species.

Impacts of Nr on freshwater ecosystems

Reactive nitrogen (Nr), including reduced (ammonium, organic N compounds) and oxidized (nitrate, nitrite) forms, play central roles in modulating and controlling (limiting) primary and secondary production and species composition in freshwater ecosystems. These include lakes, reservoirs, streams, rivers, and wetlands (Goldman, 1981; Paerl, 1982; Elser et al., 1990, 2007; Wetzel, 2001). While phosphorus has been considered the primary limiting nutrient in freshwater ecosystems (c.f. Schindler, 1971; Schindler et al., 2008), there are numerous examples where Nr plays either a primary or secondary (i.e., co-limiting) role as a limiting nutrient (Paerl, 1982; North et al., 2007; Wurtsbaugh et al., 1997; Lewis and Wurtsbaugh, 2008). In particular, oligotrophic, alpine, tropical and subtropical, and other lakes having small watersheds

Box G-1: The National Criterion for Ammonia in Fresh Water

The available data for ammonia, evaluated using the procedures described in the "Guidelines for Deriving Numerical National Water Quality Criteria for the Protection of Aquatic Organisms and Their Uses," indicate that, except possibly where an unusually sensitive species is important at a site, freshwater aquatic life should be protected if both of the following conditions are satisfied for the temperature (T) and pH of the waterbody:

1. The one-hour average concentration of total ammonia nitrogen (in mgN/L does not exceed, more than once every three years on the average, the CMC (acute criterion) calculated using the following equations. Where salmonid fish are present:

$$CMC = \frac{0.275}{1+10^{7.204-pH}} + \frac{39.0}{1+10^{pH-7.204}}$$

Or where salmonid fish are not present:

$$CMC = \frac{0.411}{1+10^{7.204-pH}} + \frac{58.4}{1+10^{pH-7.204}}$$

2A. The 30-day average concentration of total ammonia nitrogen (in mg N/L) does not exceed, more than once every three years on the average, the CCC (chronic criterion) calculated using the following equations.

When fish early life stages are present:

$$CCC = \left(\frac{0.0577}{1+10^{7.688-pH}} + \frac{2.487}{1+10^{pH-7.688}} \right) \cdot MIN(2.85\,,\ 1.45 \cdot 10^{(0.28 \cdot 25-T)})$$

When fish early life stages are absent:

$$CCC = \left(\frac{0.0577}{1+10^{7.688-pH}} + \frac{2.487}{1+10^{pH-7.688}} \right) \cdot 1.45 \cdot 10^{0.028 \cdot (25-MAX(T,7))}$$

2B. In addition, the highest 4-day average within the 30-day period should not exceed 2.5 times the CCC.

Source: U.S. EPA, 1999.

relative to the lake surface/volume, and lakes experiencing incipient stages of eutrophication, tend to be N-limited (Wetzel, 2001; Lewis and Wurtsbaugh, 2008). N limitation was illustrated for Lake Tahoe (in California and Nevada) which was highly sensitive to N enrichment during its early stages of eutrophication (Goldman, 1981; 1988). As the lake accumulated anthropogenic N inputs from both land-based runoff and atmospheric deposition within the Tahoe Basin, it began exhibiting symptoms of accelerating eutrophication, including noticeable "greening" of its formerly transparent near-shore waters and excessive epiphytic growth and fouling on its rocky bottom. Continued excessive N loading in the 1960s through 1980s has led to accelerating rates of algal primary production and a tendency to shift to more P limited conditions due to excessive N, relative to P, loading (Goldman,1988). This greater than 30-year progression to more eutrophic, and less desirable (from ecological, trophic and economic perspectives—i.e., tourism, water use) conditions has largely been spurred on by excessive N loading. Recent measures taken to reduce N inputs have been successful in reducing the lake's rate of eutrophication (Goldman, 2002). Similarly, Lake Erie, which has experienced P-driven nuisance algal blooms starting in the 1950s, is now facing excessive N loading. This is largely a result of P input restrictions, which have been enacted since the 1970's, accompanied by a lack of control on ever-increasing N loads. This shift in nutrient loading (increasing N:P) has led to a resurgence of toxic cyanobacterial blooms dominated by the non-N2 fixing genus *Microcystis,* an indicator of excessive N loading (North et al., 2007).

Numerous lakes, reservoirs, rivers, estuaries (e.g., the Gulf of Mexico), and fjords worldwide exhibit N and

P co-limitation, either simultaneously or in seasonally-shifting patterns (Dodds et al., 1989; Elser et al., 1990, 2007; Elmgren and Larsson, 2001; Forbes et al., 2008; Scott et al., 2008; Wetzel, 2001; North et al., 2007; Xu et al., 2010). For example, many reservoirs in Texas exhibit seasonal N limitation in the river-reservoir transition zone, regardless of their trophic status (Scott et al., 2009). Under these circumstances, N inputs tend to determine the spatial and temporal extent of summer nuisance algal blooms, a key symptom of degrading water quality (Dodds et al., 1989; Paerl, 2009; Xu et al., 2010). N inputs, including those from increasing levels of atmospheric deposition, impact nutrient stoichiometry, with cascading effects on nutrient limitation, productivity, and lake nutrient cycling characteristics (Elser et al., 2009). Therefore, the inputs of N play a critical role in the overall trophic response, trophic state, and water quality conditions of affected freshwater ecosystems.

In Florida lakes, algae are often limited by the availability of Nr (Kratzer and Brezonik, 1981). The most well-studied example is Lake Okeechobee, the largest lake in the Southeastern U.S., and a system that periodically displays large blooms of noxious blue-green algae. This lake has high availability of reactive P, and changes in the availability of Nr control the wax and wane of algae. In the 1980s and 1990s, blooms of algae were predominantly caused by cyanobacterial nitrogen (N2) fixer *Anabaena.* However, the most widespread recent bloom, which covered almost the entire lake surface in summer 2006, was caused by *Microcystis,* a non-N2- fixing cyanobacterium that depends on dissolved inorganic N (DIN: ammonium, nitrate, nitrite) and possibly organic N for its growth. This alga is the most common producer of toxins in Florida lakes, and it has the ability to "luxury consume" P from lake sediments and then rise through the water column, increasing its biomass to a level that largely is controlled by the amount of DIN. Because Lake Okeechobee's sediments contain massive quantities of reactive P (Havens et al., 2007), successful control of *Microcystis* blooms will require reduction in *both* P and N inputs to this lake.

In addition to the importance *total* N loads play in determining water quality status and trends, the supply rates and ratios of various Nr forms play an important role in structuring microalgal and macrophyte communities mediating freshwater primary production (Paerl, 1988; McCarthy et al., 2007, 2009; Lin et al., 2008). For example, the ratio of ammonium to oxidized N was related to the proportion of cyanobacteria composing the total phytoplankton community of Lake Okeechobee (McCarthy et al., 2009). Non-N_2-fixing cyanobacteria, such as Microcystis, are superior competitors for reduced N (Blomqvist et al., 1994), but even N_2-fixing cyanobacteria will preferentially assimilate ammonium if it is available (Ferber et al., 2004). Ammonium is the initial N form produced by recycling processes (via invertebrate excretion and bacterial mineralization), but standing concentrations often remain very low because they are assimilated rapidly. Ammonium and other reduced N forms, such as dissolved free amino acids, are more available than oxidized N forms (nitrate and nitrite) to bacteria (Vallino et al., 1996) and cyanobacteria because less energy is required to incorporate reduced N into biomass than for oxidized forms (Syrett, 1981; Gardner et al., 2004; Flores and Herrero, 2005).

Lastly, it should be pointed out that both freshwater and marine systems do not respond to nutrient inputs in isolation. These systems are hydrologically and biogeochemically connected and coupled, functioning as a freshwater to marine *continuum* (Paerl, 2009). Nutrient limitation may shift along the continuum, and eutrophication and other symptoms of N and P over-enrichment, including harmful algal blooms, hypoxia, loss of biodiversity, and food web alterations impact water quality, habitat condition, use, and sustainability of downstream waters. Therefore, excessive N loading in upstream freshwater ecosystems, ranging from the headwaters of pristine alpine streams to lowland lakes, reservoirs, and rivers can adversely affect downstream estuarine and coastal marine waters (Conley et al., 2009; Paerl, 2009). Examples of such continuum-scale impacts include such prominent systems as Chesapeake Bay, Albemarle-Pamlico Sound, Florida Bay, Mississippi River plume (Gulf of Mexico), Baltic Sea, and Coastal North Sea (Elmgren and Larsson, 2001; Boesch et al., 2001; Paerl, 2009).

Impact of nitrogen on wetlands

In this section, the possible impact of reactive nitrogen on wetlands is discussed. In Chapter 5 considerable attention is devoted to the subject of wetlands serving as effective nitrogen sinks. There are about 110 million ha of wetlands in the U.S., with more than half of those in the state of Alaska (Mitsch et al., 2009). Of those wetlands, roughly 97% are inland (and mostly freshwater) and 3% are estuarine (and mostly saline). Of the total wetlands, approximately half (166 million acres or 55 million ha) are peatlands, which, by their nature as low-nutrient systems, are most susceptible to nitrogen loadings, either from the atmosphere or from rivers and streams. More than any other ecosystem, wetlands are central to the cycling of nitrogen because they have both aerobic and anaerobic conditions that allow for a wide variety of important nitrogen processes, not the least of which is denitrification.

Wetlands are similar to lakes and streams and any other ecosystem in that their productivity is limited by nutrient availability. But with wetlands the hydrology limits or enhances productivity as well (Mitsch and Gosselink, 2007). The addition of excessive nutrients to wetlands, while often done purposefully when the wetlands are so-called treatment wetlands (Kadlec and

Wallace, 2009), can cause vegetation shifts and decreases in plant diversity. Verhoeven et al. (2006) suggested 4.5 g-N m^{-2} yr^{-1} as critical loading rate of nitrogen, generally from atmospheric sources, for peat-dominated wetlands. Morris (1991) had suggested that bogs and fens generally had loading rates of 1 to 6 g-N m^{-2} yr^{-1} respectively. Thus Verhoeven et al. (2006) were suggesting that wetlands should not be loaded beyond what is currently occurring in fen peatlands. These limitations do not apply for most mineral soil wetlands, particularly those connected to streams and rivers. Most freshwater and tidal marshes have nitrogen loading rates closer to 60 g-N m^{-2} yr^{-1} and they maintain a reasonably high and sustainable productivity. Using a sustainable rate of nitrogen retention as a measure, Mitsch and Jørgensen (2004) suggest a range of nitrogen retention rate of 10-20 g-N m^{-2} yr^{-1} for wetlands to maintain their biodiversity while being nitrogen sinks at the same time. Overall, this is a fruitful direction for wetland research to determine the assimilative capacity of wetlands for nutrients, including nitrogen, while not surpassing a limit that will dramatically change the wetland's structure and function.

Impacts of Nr on coastal systems

Mitsch et al. (2001) suggest that streams and rivers themselves are not always as much affected by nutrient loading as are lakes, wetlands, coastal areas, and other lentic bodies of water. However, in most cases, these nutrient-enriched waterways flow to the sea, with eutrophication of coastal waters the unfortunate result. This problem now occurs regularly throughout the world (World Resources Institute, 2008), in locations such as the Gulf of Mexico (Rabalais et al., 1996), the Baltic Sea (Larson et al., 1985), and the Black Sea (Tolmazin, 1985).

During the past century, following large-scale use of synthetic N fertilizers in agriculture, rapid expansion of industrial and transportation-related fossil fuel combustion and coastal urbanization, humans have significantly altered the balance between "new" N inputs and N losses in the marine environment (Codispoti et al., 2001; Galloway and Cowling, 2002). During this time frame, terrestrial discharge and atmospheric N emissions have increased 10-fold (Howarth et al., 1996; Holland et al., 1999). This number keeps growing as human development continues to expand in coastal watersheds (Vitousek et al., 1997a,b).

Researchers have long recognized this growing imbalance, especially in estuarine and coastal waters where anthropogenically-derived N over-enrichment has fueled accelerated primary production, or "cultural" eutrophication (Vollenweider et al., 1992; Nixon, 1995). Eutrophication is a condition where nutrient-enhanced primary production exceeds the ability of higher ranked consumers and organic matter-degrading microbes to consume and process it. D'Elia (1987) characterized this condition as "too much of good thing" or over-fertilization of N-limited marine ecosystems with "new" N, the bulk of it being anthropogenic (Howarth et al., 1996; Vitousek et al., 1997a,b; Galloway and Cowling, 2002). Symptoms of N-driven eutrophication vary: from subtle increases in plant production to changes in primary producer community composition; to rapidly accelerating algal growth, visible discoloration or blooms, losses in water clarity, increased consumption of oxygen, dissolved oxygen depletion (hypoxia), which is stressful to resident fauna and flora; to, in the case of total dissolved oxygen depletion (anoxia), elimination of habitats (Paerl, 1988, 1997; Diaz and Rosenberg, 1995; Rabalais and Turner, 2001). Other effects include submerged aquatic vegetation (SAV) losses, possible impacts on tidal wetland health, and disruption of estuarine food chain dynamics that may favor an imbalance towards lower trophic levels (e.g., jellyfish).

Anthropogenic or cultural eutrophication has been closely linked to population densities in coastal watersheds (Peierls et al., 1991; Nixon, 1995; Vitousek et al., 1997a,b). Primary sources of N enrichment include urban and agricultural land uses as well as wastewater treatment plants, many of which have not been designed to remove N. A significant, and in many instances increasing, proportion of "new" N input can also be attributed to remote sources residing in airsheds. Delivery routes can also be complex, especially when via subsurface aquifers outside the immediate watershed, which can confound source definition and create long delays in delivery and management response (Paerl, 1997; Jaworski et al., 1997; Galloway and Cowling, 2002; Paerl et al., 2002).

The availability of N controls primary production in much of the world's estuarine, near-shore coastal, and open-ocean waters (Dugdale, 1967; Ryther and Dunstan, 1971; Nixon, 1995; Paerl, 1997; Boesch et al., 2001). As previously discussed, nitrogen can also play a role as either a primary or secondary limiting nutrient in freshwater environments, especially large lakes (e.g., Lake Tahoe, Lake Superior). As such, the fertility of these waters is often closely controlled by N inputs, which are provided either internally by regeneration of pre-existing N and biologically-fixed atmospheric N2, or supplied externally (i.e. "new" N) as combined N sources delivered via surface runoff, sub-surface groundwater, or atmospheric deposition.

The extent to which accelerated N loading promotes eutrophication and its symptoms varies greatly among marine ecosystems. Receiving waters exhibit variable sensitivities to N and other nutrient [phosphorus (P), iron (Fe), and silica (Si)] loads that are controlled by their size, hydrologic properties (e.g., flushing rates and residence times), morphologies (depth, volume), vertical mixing characteristics, geographic and climatic regimes and conditions. In addition, the magnitude and distribution of N in relation to other nutrient loads can vary substantially. In waters receiving very high N loads relative to requirements for sustaining primary and secondary production, other nutrient limitations may develop. This appears to be the case in coastal waters downstream of rivers draining agricultural regions that are enriched in N.

On the ecosystem level, estuarine and coastal waters exhibit individualistic responses to N loads over seasonal and longer (multi-annual, decadal) time scales. The degree to which these systems are exposed to freshwater discharge, tidal exchange, and vertical mixing is critical for determining how they respond to specific N loads (Vollenweider et al., 1992; Nixon, 1995; Cloern, 1999, 2001; Valdes-Weaver et al., 2006; Paerl et al., 2007). Another variable is the manner in which N loading takes place, which may range from acute pulsed events such as storms and associated flooding, to longer-term gradual (chronic) increases in N loading associated with more predictive seasonal, annual and inter-annual hydrologic cycles. There are striking contrasts in ecosystem response to N inputs that reflect a range in physical (hydrodynamic, optical) and climatic conditions (Cloern, 1999, 2001). Examples include contrasts between strong tidally-driven estuarine systems, such as Delaware Bay and San Francisco Bay, and non-tidal, lagoonal systems, such as North Carolina's Pamlico Sound and Texas's Luguna Madre, or semi enclosed coastal systems, such as Florida Bay and the Long Island Sound (Bricker et al., 1999; Valdes-Weaver et al., 2006; Paerl et al., 2007).

Externally-supplied N comes in various forms, including organic N and inorganic reduced (NH_3 and NH_4^+ ion) and oxidized (NO_3^-) N, all of which are potentially available to support new production and eutrophication. Laboratory experiments on phytoplankton isolates and bioassays with natural phytoplankton communities have indicated that these contrasting forms may be differentially and preferentially utilized, indicating that, depending on composition of the affected phytoplankton community, some forms are more reactive than others (Collos, 1989; Stolte et al., 1994; Riegman, 1998). Phytoplankton community composition can also be altered by varying proportions and supply rates of different forms of N (Dortch, 1990; Stolte et al., 1994; Harrington, 1999; Pinckney et al., 1999; Piehler et al., 2002). Monitoring and research on dissolved organic N inputs and their effects should be conducted in receiving streams, rivers, lakes, estuarine, and coastal waters, since there is evidence that these compounds can be utilized by phytoplankton, including harmful bloom species (Paerl, 1988; Antia et al., 1991; Carlsson and Granéli, 1998; Gilbert et al., 2006). In addition, specific N compounds may interact with light availability, hydrodynamics and other nutrients, most notably P, Si, Fe, and trace metals, to influence phytoplankton community growth rates and composition (Harrison and Turpin, 1982; Smith, 1990; Dortch and Whitledge, 1992).

Over the past 25 years, there has been a growing recognition of cultural eutrophication as a serious problem in coastal estuaries (NRC, 2000). Globally, Selman et al. (2008) have reported "Of the 415 areas around the world identified as experiencing some form of eutrophication, 169 are hypoxic and only 13 systems are classified as 'systems in recovery."Comprehensive surveys of U.S. estuaries have been conducted by the National Oceanic and Atmospheric Administration (NOAA) as part of the National Estuarine Eutrophication Assessments (NEEA) in 1999 and 2004 (Bricker et al., 1999, 2007). The most recent report, released in 2007 (Bricker et al., 2007) focused on nutrient enrichment and its manifestations in the estuarine environment and relies on participation and interviews of local experts to provide data for the assessment. Among the key findings for nearly 100 assessed U.S. estuaries were that eutrophication is a widespread problem, with the majority of assessed estuaries showing signs of eutrophication—65% of the assessed systems, representing 78% of assessed estuarine area, had moderate to high overall eutrophic conditions. The most common symptoms of eutrophication were high spatial coverage and frequency of elevated chlorophyll *a* (phytoplankton)—50% of the assessed estuaries, representing 72% of assessed area, had a high chlorophyll *a* rating.

Further field evaluations by EPA and state and university collaborators under the National Coastal Assessment (NCA) used probabilistic monitoring techniques. The NCA National Coastal Condition Reports (NCCR) (U.S. EPA, 2001a, 2004, 2006b) are more closely related to nutrient enrichment assessments, especially for manifestations of nutrient enrichment such as hypoxia, nuisance algal blooms, and general habitat degradation. The last comprehensive national NCCR was published in 2004 (U.S. EPA, 2004) with a more recent assessment focused on 28 National Estuary Program estuaries published in 2007 (U.S. EPA, 2006). The 2004 NCCR included an overall rating of "fair" for estuaries, including the Great Lakes, based on evaluation of more than 2,000 sites. The water quality index, which incorporates nutrient effects primarily as chlorophyll-*a* and dissolved oxygen impacts, was also rated "fair" nationally. Forty percent of the sites were rated "good" for overall water quality, while 11% were "poor" and 49% "fair."

Attainment of water quality management goals and standards for coastal systems

Estuarine systems, where bio-available Nr is more likely to be the limiting nutrient, are most often susceptible to Nr enrichment (Paerl, 1997; Boesch et al., 2001). Defining single-number criteria for nutrients or related indicators representative of undesirable levels of productivity (e.g., chlorophyll-*a*) is difficult, even using the ecoregional approach recommended by EPA. State managers more often use the formal TMDL process or collaborative estuarine management plans to set site- or estuary-specific N management targets to meet existing, related water quality criteria (e.g., dissolved O_2 or chlorophyll *a*). Some of the more prominent efforts and targets for nitrogen control are summarized in Table G-1.

These targets all exceed nitrogen load reduction goals that the Committee has found to be readily achievable using existing technology and management authority (i.e., less than 25% from specific source categories). Some sources of nitrogen loading to the estuaries in Table G-1 pose greater management challenges and the expectation is that reductions in Nr loadings to estuaries would cumulatively be less than 25%. This suggests that efforts will be needed to enable even greater nitrogen load reductions in the future. Many of the management actions the Committee has proposed in this report would require substantive changes in national programs, regulatory authority, management technologies and societal demands to be accomplished. This is a nutrient management concern that state managers are well aware of as they develop TMDLs and management plans that range above attainment potential, not only for Nr but more frequently for other pollutants that are predominately nonpoint source and stormwater loaded (including atmospheric source contributions).

The Chesapeake Bay Program, for example, is a model for Nr and P management in many ways. Considerable resources were committed, and many BMPs were implemented. Yet despite regional efforts and commitments from all watershed states, and more funding than any other estuary program is likely to see, management targets have not been met, and recent data (2007) reveal the occurrence of a severe hypoxic episode. Concerns over the slow progress in restoring the Chesapeake Bay led to the issuance of an Executive Order on May 15, 2009, establishing a Federal Leadership Committee led by the EPA to develop and implement a plan to restore the Bay in collaboration with state agencies (*Federal Register* 74(93): 23097-23104). Similarly, the adoption of the Long Island Sound TMDL, which was driven by the presence of reactive nitrogen (see Box G-2), sets an implementation plan that could attain Connecticut and New York dissolved oxygen criteria, but only if “alternative technologies” such as mechanical aeration of the Sound or biological harvesting of nutrients, are used.

Table G-1: Estuaries with nitrogen management plans or TMDLs and percent nitrogen load reduction targets

Estuary	Nitrogen Load Reduction Target	TMDL or Plan
Casco Bay, Maine	45%	Plan
Chesapeake Bay	>40%	Plan
Northern Gulf of Mexico		
Mississippi Plume Region	45%	Plan
Long Island Sound	60% for CT & NY sources	TMDL
Neuse River Estuary, NC	30%	TMDL
Tampa Bay, FL	Maintain TN (total nitrogen) load at 1992-1994 levels	TMDL & Plan

Box G-2: Long Island Sound Total Maximum Daily Load: Focus on Reactive Nitrogen

A TMDL sets a goal for reducing the load of a specific pollutant that is causing impairment to a waterbody. In the case of Long Island Sound, the impairment constitutes low concentrations of dissolved O_2 that violate both Connecticut's and New York's water quality standards. Nitrogen has been identified as the pollutant that causes substandard levels of dissolved oxygen in Long Island Sound and, accordingly, Connecticut's and New York's environmental agencies have developed a TMDL that assigns nitrogen reductions from both point sources (the wasteload allocation or WLA) and nonpoint sources (the load allocation or LA) in their respective states to meet the established 58.5% reduction of anthropogenic sources.

The Long Island Sound TMDL is set at 23,966 tons of N/year, which represents a 23,834 ton/year reduction from the total baseline (anthropogenic + natural sources considered) of 47,788 tons/year from Connecticut and New York only. Most of that N load comes from point sources – POTWs (publicly owned treatment works) and CSOs (combined sewer overflows) – accounting for 38,899 tons/yr of the total N load from the two states, or 81% of the load. For that reason, the focus has been on managing point sources, although attainment of water quality standards will require more widespread reductions from atmospheric deposition, stormwater, and nonpoint sources, and from other watershed states north of Connecticut.

Connecticut and New York have some flexibility in the apportionment of those reductions between the WLA and the LA, but must have completed 40% of the required reductions by 2004, 75% by 2009, and 100% by 2014 when the final TMDL will be met. However, the TMDL is presently undergoing revision to incorporate findings from a new model of Long Island Sound, and to reflect changes in dissolved O_2 criteria in both states. The revised TMDL will likely require more aggressive reductions of nitrogen to meet dissolved O_2 criteria and may formalize targets for upstream state contributions and atmospheric deposition.

Appendix H:
Nr Saturation and Ecosystem Function

There are limits to how much plant growth can be increased by N fertilization. At some point, when the natural N deficiencies in an ecosystem are fully relieved, plant growth becomes limited by availability of other resources such as phosphorus, calcium, or water and the vegetation can no longer respond to further additions of Nr. In theory, when an ecosystem is fully Nr-saturated and its soils, plants, and microbes cannot use or retain any more, all new Nr deposits will be dispersed to streams, groundwater, and the atmosphere. Nr saturation has a number of damaging consequences for the health and functioning of ecosystems. These impacts first became apparent in Europe almost three decades ago when scientists observed significant increases in nitrate concentrations in some lakes and streams and also extensive yellowing and loss of needles in spruce and other conifer forests subjected to heavy Nr deposition. In soils, most notably forest soils because of their natural low pH, as NH_4^+ builds up it is converted to nitrate by bacterial action, a process that releases hydrogen ions and contributes to soil acidification. The buildup of NO_3^- enhances emissions of nitrous oxides from the soil and also encourages leaching of highly water-soluble NO_3^- into streams or groundwater. As negatively charged NO_3^- seeps away, positively charged alkaline minerals such as calcium, magnesium, and potassium are carried along. Thus, soil fertility is decreased by greatly accelerating the loss of calcium and other nutrients that are vital for plant growth. As calcium is depleted and the soil acidified, aluminum ions are mobilized, eventually reaching toxic concentrations that can damage tree roots or kill fish if the aluminum washes into streams (Vitousek et al., 1997a,b).

Forests, grasslands, and wetlands vary substantially in their capacity to retain added nitrogen. Interacting factors that are known to affect this capacity include soil texture, degree of chemical weathering of soil, fire history, rate at which plant material accumulates, and past human land use. However, we still lack a fundamental understanding of how and why N-retention processes vary among ecosystems, much less how they have changed and will change with time and climate change (Clark and Tilman, 2008).

An overarching impact of excess Nr on unmanaged terrestrial ecosystems is biodiversity loss. In North America, dramatic reductions in biodiversity have been created by fertilization of grasslands in Minnesota and California. In England, N fertilizers applied to experimental grasslands have led to similarly increased dominance by a few N-responsive grasses and loss of many other plant species. In formerly species-rich heathlands across Western Europe, Nr deposition has been blamed for great losses of biodiversity in recent decades, with shallow soils containing few alkaline minerals to buffer acidification (Vitousek et al., 1997a,b; Bobbink et al., 2010).

Losses of biodiversity driven by Nr deposition can in turn affect other ecological processes. Experiments in Minnesota grasslands showed that in ecosystems made species-poor by fertilization, plant productivity was much less stable in the face of a major drought. Even in non-drought years, the normal vagaries of climate produced much more year-to-year variation in the productivity of species-poor grassland plots than in more diverse plots (Vitousek et al., 1997a,b).

References

Abbott, P., C. Hurt, and W. Tyner. 2008. *What's Driving Food Prices? Farm Foundation Issues Report.* http://farmfoundation.org/webcontent/Farm-Foundation-Issue-Report-Whats-Driving-Food-Prices-404.aspx?z=na&a=404 [accessed August 23, 2010]

Adviento-Borbe, M.A.A., M.L. Haddix, D.L. Binder, D T. Walters and A. Dobermann. 2006. Soil greenhouse gas fluxes and global warming potential in four high-yielding maize systems. *Global Change Biology* 13:1972–1988.

Alexander, R.B., Smith, R.A. and G.E. Schwarz. 2000. Effect of stream channel size on the delivery of nitrogen to the Gulf of Mexico. *Nature* 403:758-761.

Alexander, R B., R.A. Smith, G.E. Schwarz, E.W. Boyer, J.V. Nolan, and J.W. Brakebill. 2008. Differences in sources and recent trends in phosphorous and nitrogen delivery to the Gulf of Mexico from the Mississippi and Atchafalaya River Basins. *Environmental Science and Technology* 42(3): 822-830.

Aneja, V.P., W.P. Robarge, L.J. Sullivan, T.C. Moore, T.E. Pierce, C. Geron and B. Gay. 1996. Seasonal variations of nitric oxide flux from agricultural soils in the Southeast United States. *Tellus,* 48B:626-640.

Aneja, V.P., J.P. Chauhan, and J.T. Walker. 2000. Characterization of atmospheric ammonia emissions from swine waste storage and treatment lagoons. *Journal of Geophysical Research-Atmospheres* 105:11535-11545.

Aneja, V.P., P.A. Roelle, G.C. Murray, J. Southerland, J.W. Erisman, D. Fowler, W.A.H. Asman, and N. Patni. 2001. Atmospheric nitrogen compounds: II. Emissions, transport, transformation, deposition and assessment. *Atmospheric Environment* 35:1903–1911.

Aneja, V.P., D.R. Nelson, P.A. Roelle, J.T. Walker, and W. Battye. 2003. Agricultural ammonia emissions and ammonium concentrations associated with aerosols and precipitation in the southeast United States. *Journal of Geophysical Research-Atmospheres* 108:4152.

Aneja, V.P., W.H. Schlesinger, D. Niyogi, G. Jennings, W. Gilliam, R.E. Knighton, C.S.Duke, J. Blunden, and S. Krishnan. 2006a. Emerging national research needs for agricultural air quality. *Eos, Transactions, American Geophysical Union* 87(3):25-29.

Aneja, V.P., W.H. Schlesinger, R.E. Knighton, G. Jennings, D. Niyogi, W. Gilliam, and C. Duke. 2006b. *Proceedings, Workshop on Agricultural Air Quality: State of the Science,* ISBN 0-9669770-4-1 [Available at: www.ncsu.edu/airworkshop/]

Aneja, V.P., S.P. Arya, I.C. Rumsey, D.S. Kim, K. Bajwa, and C.M. Williams. 2008a. Characterizing ammonia emissions from swine farms in Eastern North Carolina: Reduction of emissions from water-holding structures at two candidate superior technologies for waste treatment. *Atmospheric Environment* 42(14):3291-3300.

Aneja, V.P., J. Blunden, K. James, W.H. Schlesinger, R. Knighton, W. Gilliam, D. Niyogi, and S. Cole. 2008b. Ammonia Assessment from Agriculture: U.S. Status and Needs. *Journal of Environmental Quality* 37:515-520.

Aneja, V.P., J. Blunden, P.A. Roelle, W.H. Schlesinger, R. Knighton, W. Gilliam, D. Niyogi, G. Jennings, and Cliff Duke. 2008c. Workshop on agricultural air quality: State of the science. *Atmospheric Environment* 42(14):3195-3208.

Aneja, V.P., W.H. Schlesinger, and J.W. Erisman. 2008d. Farming pollution. *Nature Geoscience* 1:409-411.

Aneja, V.P., W.H. Schlesinger, and J.W. Erisman. 2009. Effects of Agriculture upon the Air Quality and Climate: Research, Policy and Regulations. *Environmental Science and Technology* 43(12):4234-4240.

Antia, N., P. Harrison and L. Oliveira. 1991. The role of dissolved organic nitrogen in phytoplankton nutrition, cell biology and ecology. *Phycologia* 30:1-89.

Appel, T. 1994. Relevance of soil N mineralization, total N demand of crops, and efficiency of applied N for fertilizer recommendations for cereals-Theory and application. Z. *Pflanzenernahr Bodenk* 157:407-414.

Archibald, A.T., M.E. Jenkin, and D.E. Shallcross. 2010. An isoprene mechanism intercomparison. *Atmospheric Environment,* 44:5356-5364.

Arogo, J., P.W. Westerman, A.J. Heber, W.P. Robarge, and J.J. Classen J J. 2006. Ammonia emissions from animal feeding operations. In *Animal Agriculture and the Environment: National Center for Manure and Animal Waste Management White Papers,* eds. J.M. Rice, D.F. Caldwell, and F.J. Humenik, 41–88. ASABE, St. Joseph, MI

Asman, W.A.H. 1994. Emission and deposition of ammonia and ammonium. *Nova Acta Leopold* 228:263-297.

Aulakh, M.S., J.W. Doran, and A.R. Mosier. 1992. Soil denitrification, significance, measurement and effects of management. *Advances in Soil Science.* 18:1-57.

Augspurger, T., A.E. Keller, M.C. Black, W.G. Cope and J. Dwyer. 2003. Water quality guidance for protection of freshwater mussels (Unionidae) from ammonia exposure. *Environmental Toxicology and Chemistry* 22(11):2569-2575.

Baek, B.H., V.P. Aneja, and Quansong Tong. 2004a. Chemical coupling between ammonia, acid gases, and fine particles. *Environmental Pollution* 129:89-98.

Baek, B.H., and V.P. Aneja. 2004b. Measurement and analysis of the relationship between ammonia, acid gases, and fine particles in Eastern North Carolina. *Journal of Air and Waste Management Association.* 54:623-633.

Baker, J.M., T.E. Ochsner, R.T. Venterea and T.J. Griffis. 2007. Tillage and soil carbon sequestration, What do we really know? *Agriculture, Ecosystems and Environment* 118:1-5.

Barber, N.L. 2009. *Summary of Estimated Water Use in the United States in 2005.* U.S. Geological Survey Fact Sheet. 2009, 4 p. U.S. Geological Survey, Washington, DC.

Bare, J.C., G. A. Norris, D. W. Pennington, and T. McKone. 2003. TRACI: The tool for the reduction and assessment of chemical and other environmental impacts. *Journal of Industrial Ecology* 6: 49-78.

Batie, S.S. 2008. Wicked problems and applied economics. *American Journal of Agricultural Economics* 90:1176-1191.

Battye, R., W. Battye, C. Overcash, and S. Fudge. 1994. *Development and selection of ammonia emission factors.* Final Report. EC/R Inc. for U.S. EPA, EPA/600/R-94/190.

Beard, J.B., and R.L. Green. 1994. The role of turfgrassses in environmental protection and their benefits to humans. *Journal of Environmental Quality* 23:452-460.

Beard, J.B. and Kenna, M.P. (Eds.). 2008. *Water Quality and Quantity Issues for Turfgrasses in Urban Landscapes.* Council for Agricultural Science and Technology. Ames, Iowa, U.S. 298 p.

Bequette, B.J, M.D. Hanigan, and H. Lapierre. 2003. Mammary uptake and metabolism of amino acids by lactating ruminants. In *Amino Acids in Animal Nutrition, 2nd edition,* ed. J.P.F. D'Mello, 347-365. CAB International, Oxon, UK.

Bicudo, J.R., C.J. Clanton, D.R. Schmidt, W. Powers, L.D. Jacobson, and C.L. Tengman. 2004. Geotextile covers to reduce odor and gas emissions from swine manure storage ponds. *Applied Engineering in Agriculture* 20:65-75.

Birch, M.B.L., B.M. Gramig, W.R. Moomaw, O.C. Doering, III, and C.J. Reeling. 2011. Why metrics matter: Evaluating policy choices for reactive nitrogen in the Chesapeake Bay Watershed. *Environmental Science and Technology* 45(1): 168-174. [available at: http://pubs.acs.org/doi/abs/10.1021/es101472z]

Birdsey, Richard, A. 1992. *Carbon Storage and Accumulation in United States Forest Ecosystems.* U.S. Department of Agriculture Forest Service General Technical Report WO-59, U.S. Forest Service Northeastern Forest Experiment Station, Radnor, PA [available at: http://nrs.fs.fed.us/pubs/gtr/gtr_wo059.pdf]

Blanco-Canqui, H., and R. Lal. 2008. No-tillage and soil-profile carbon sequestration: An on-farm assessment. *Soil Science Society of America Journal* 72:693-701.

Bleken M.A., and L.R. Bakken.1997. The nitrogen cost of food production: Norwegian Society. *AMBIO: A Journal of the Human Environment* 26:134-142

Bleken, M.A., H. Steinshamn, and S. Hansen. 2005. High nitrogen costs of dairy production in Europe: Worsened by intensification. *AMBIO: A Journal of the Human Environment* 34:598-606.

Blomqvist, P., A. Pettersson, and P. Hyenstrand. 1994. Ammonium-nitrogen: a key regulatory factor causing dominance of non-nitrogen-fixing cyanobacteria in aquatic systems. *Archiv für Hydrobiologie* 132:141-164.

Bloomer, B.J., J.W. Stehr, C.A. Piety, R.J. Salawitch, and R.R. Dickerson. 2009. Observed relationships of ozone air pollution with temperature and emissions. *Geophysical Research Letters* 36, doi:10.1029/2009GL037308

Blowes, D.W., W.D. Robertson, C.J. Ptacek, and C. Merkley. 1994. Removal of agricultural nitrate from tile-drainage effluent water using in-line bioreactors. *Journal of Contaminant Hydrology* 15(3):207-221.

Bobbink, R., Hicks, K., Galloway, J., Spranger, T., Alkemade, R., Ashmore, M., Bustamante, M., Cinderby, S., Davidson, E., Dentener, F., Emmett, B., J-W Erisman, M. Fenn, F. Gilliam, A. Nordin, L. Pardo, and W. de Vries. 2010. Global assessment of nitrogen deposition effects on terrestrial plant diversity. *Ecological Applications* 20(1): 30–59.

Boesch, D.F., E. Burreson, W. Dennison, E. Houde, M. Kemp, V. Kennedy, R. Newell, K. Paynter, R. Orth, and R. Ulanowicz. 2001. Factors in the decline of coastal ecosystems. *Science* 293:629-638.

Booth, M., and C. Campbell. 2007. Spring Nitrate Flux in the Mississippi River Basin: A Landscape Model with Conservation Applications. *Environmental Science and Technology* 41:5410-5418.

Boyer, E.W., C.L. Goodale, N.A. Jaworski, and R.W. Howarth. 2002. Anthropogenic nitrogen sources and relationships to riverine nitrogen export in the northeastern U.S. *Biogeochemistry* 57:137-169.

Bricker, S.B., C.G. Clement, D.E. Pirhalla, S.P. Orlando and D.R.G. Farrow. 1999. *National Estuarine Eutrophication Assessment: Effects of Nutrient Enrichment in the Nation's Estuaries.* NOAA, National Ocean Service Special Projects Office and the National Centers for Coastal Ocean Science, Silver Spring, MD. 71 p.

Bricker, S., B. Longstaff, W. Dennison, A. Jones, K. Boicourt, C. Wicks, C, and J. Woerner. 2007. *Effects of Nutrient Enrichment in the Nation's Estuaries: A Decade of Change.* NOAA Coastal Ocean Program Decision Analysis Series No. 26. National Centers for Coastal Ocean Science. Silver Spring, MD.

Brook, J.R., F. Di-Giovanni, S. Cakmak, and T.P. Meyers. 1997. Estimation of dry deposition velocity using inferential models and site-specific meteorology - Uncertainty due to siting of meteorological towers. *Atmospheric Environment* 31: 3911-3919.

Brook R.D., J.R. Brook, and S. Rajagopalan. 2003. Air pollution: the "Heart" of the problem. *Current Hypertension Reports* 5(1):32-39.

Brunekreef B., N.A.H. Janssen, J.J. de Hartog J.J., M. Oldenwening, K. Meliefste, and G. Hoek. 2005. Personal, indoor, and outdoor exposures to PM$_{2.5}$ and its components for groups of cardiovascular patients in Amsterdam and Helsinki. *Research Report Health Effects Institute* 2005: Jan (127): 1-70

Burgholzer, R. 2007. *Report of a Workshop on Understanding Fertilizer Sales and Reporting Information,* May 1, 2007. U.S. EPA Chesapeake Bay Program Scientific and Technical Advisory Committee and the Agricultural Nutrient Reduction Workgroup. U.S. EPA, Washington, DC [available at: www.chesapeake.org/stac/Pubs/FertilizerReport.pdf]

Butler, T.J., G.E. Likensa, F.M. Vermeylenc, and B.J.B. Stu. 2005. The impact of changing nitrogen oxide emissions on wet and dry nitrogen deposition in the northeastern USA. *Atmospheric Environment* 39: 4851-4862.

Carlsson, P., and E. Granéli. 1998. Utilization of dissolved organic matter (DOM) by phytoplankton including harmful species. In *Physiological Ecology of Harmful Algal Blooms,* eds. Anderson, D.M., Cembella A.D. and Hallegraeff, G.M., 509-524. Springer-Verlag, Heidelburg.

Canchi, D., P. Bala, and O. Doering. 2006. *Market Based Policy Instruments in Natural Resource Conservation.* Report for the Resource Economics and Social Sciences Division, Natural Resources Conservation Service, U.S. Department of Agriculture, Washington, D.C.

Cassman, K.G. 1999. Ecological intensification of cereal production systems: Yield potential, soil quality, and precision agriculture. *Proceedings of the National Academy of Sciences of the United States of America* 96: 5952-5959.

Cassman, K.G., D.C. Bryant, A. Fulton, and L.F. Jackson. 1992. Nitrogen supply effects on partitioning of dry matter and nitrogen to grain of irrigated wheat. *Crop Science* 32:1251-1258.

Cassman, K.G., A.D. Dobermann, and D.T. Walters. 2002. Agroecosystems, N-Use Efficiency, and N Management. *AMBIO: A Journal of the Human Environment* 31:132-140.

Cassman, K.G., A. Dobermann, D.T. Walters, and H. Yang. 2003. Meeting cereal demand while protecting natural resources and improving environmental quality. *Annual Review of Environment and Resources* 28:315-358.

Castellanos, P., W.T. Luke, P. Kelley, J.W. Stehr, S.H. Ehrman, and R.R. Dickerson. 2009. Modification of a commercial cavity ring-down spectroscopy NO_2 detector for enhanced sensitivity. *Review of Scientific Instruments* 80(11):113107.

Chameides, W.L., and J.C.G. Walker. 1973. A photochemical theory of tropospheric ozone. *Journal of Geophysical Research* 78:8751-8760.

Chesapeake Bay Program. 2003a. *Economic Analyses of Nutrient and Sediment Reduction Actions to Restore Chesapeake Bay Water Quality.* U.S. Environmental Protection Agency, Region III, Chesapeake Bay Program Office, Annapolis, MD.

Chesapeake Bay Program. 2003b. *Setting and Allocating the Chesapeake Bay Basin Nutrient and Sediment Loads: The Collaborative Process, Technical Tools and Innovative Approaches;* U.S. Environmental Protection Agency, Region III, Chesapeake Bay Program Office: Annapolis, MD

Chesapeake Bay Scientific Technical Advisory Committee. 2007. *Understanding Fertilizer Sales and Reporting Information.* Workshop Report, Frederick, Maryland. Chesapeake Bay Program (U.S.) Scientific and Technical Advisory Publication 07-004 U.S. Environmental Protection Agency, Region III, Chesapeake Bay Program Office, Annapolis, MD.

Civerolo, K.L., and R.R. Dickerson. 1998. Nitric oxide soil emissions from tilled and untilled cornfields. *Agricultural and Forest Meteorology* 90:307-311.

Clark, C.M., and D. Tilman. 2008. Loss of plant species after chronic low-level nitrogen deposition to prairie grasslands. *Nature* 451:712-715.

Cloern, J.E. 1999. The relative importance of light and nutrient limitation of phytoplankton growth: a simple index of coastal ecosystem sensitivity to nutrient enrichment. *Aquatic Ecology* 33:3-16.

Cloern, J.E. 2001. Our evolving conceptual model of the coastal eutrophication problem. *Marine Ecology Progress Series* 210:223-253.

Codispoti, L.A., J.A, Brandes, J.P., Christensen, A.H. Devol, S.W.A. Naqvi, H.W. Paerl, and T. Yoshinari. 2001. The oceanic fixed nitrogen and nitrous oxide budgets: Moving targets as we enter the anthropocene? *Scientia Marina* 65(2):85-105.

Cohen, M. 2008. EPA, personal email communication, Nov. 12, 2008.

Collos, Y. 1989. A linear model of external interactions during uptake of different forms of inorganic nitrogen by microalgae. *Journal of Plankton Research* 11:521-533.

Conley, D.J., H.W. Paerl, R.W. Howarth, D.F. Boesch, S.P. Seitzinger, K.E. Havens, C. Lancelot, and G.E. Likens. 2009. Controlling eutrophication: Nitrogen and phosphorus. *Science* 323:1014-1015.

Connecticut Department of Environmental Protection (CTDEP). 2007. Nitrogen Removal Projects Financed by the CWF through 2006. Provided by Iliana Ayala, June 13, 2007.

Cooper, N.L., J.R. Bidwell, and D.S. Cherry. 2005. Potential effects of Asian clam *(Corbicula fluminea)* die-offs on native freshwater mussels (Unionidae) II: porewater ammonia. *Journal of the North American Benthological Society* 24(2):381-394.

Costanza, R., R. d'Arge, R. de Groot, S. Farber, M. Grasso, B. Hannon, S. Naeem, K. Limburg, J. Paruelo, R.V. O'Neill, R. Raskin, P. Sutton and M. van den Belt. 1997. The value of the world's ecosystem services and natural capital. *Nature* 387:253-260.

Cowling, E.B. 1989. Recent changes in chemical climate and related effects on forests in North America and Europe. *AMBIO: A Journal of the Human Environment* 18:167-171.

Cowling, E.B., D.S. Shriner, J.E. Barnard, A.A. Lucier, A.H. Johnson, and A.R. Kiester. 1990. Airborne chemicals and forest health in the United States. Volume B, pp. 25-36. In *Proceedings of the International Union of Forest Research Organizations.* Montreal, Canada.

Cowling, E.B., J.N. Galloway, C.S. Furiness, M.C. Barber, T. Bresser, K. Cassman, J.W. Erisman, R. Haeuber, R.W. Howarth, J. Melillo, W. Moomaw, A. Mosier, K. Sanders, S. Seitzinger, S. Smeulders, R. Socolow, D. Walters, F. West, and Z. Zhu. 2002. Optimizing nitrogen management in food and energy production and environmental protection: summary statement from the Second International Nitrogen Conference. In *Proceedings of the 2nd International Nitrogen Conference on Science and Policy,* eds. J. Galloway, E. Cowling, J. Erisman, J, Wisniewsky, and C. Jordan. *The Science World* 1(52):1-9.

Crampton, W.G., G.A. Stenback, B.A. Miller, and M.J. Helmers. 2006. *Potential Benefits of Wetland Filters for Tile Drainage Systems: Impact on Nitrate Loads to Mississippi River Subbasins.* U.S. Department of Agriculture Project Number 10W06682

Crosley, D.R. 1996. NO_y blue ribbon panel. *Journal of Geophysical Research-Atmospheres* 101:2049-2052.

Crutzen, P.J. 1973. A discussion of the chemistry of some minor constituents in the stratosphere and troposphere. *Pure and Applied Geophysics* 106:1385-1399.

Crutzen, P.J. 1974. Photochemical reactions initiated by and influencing ozone in unpolluted tropospheric air. *Tellus* 26:47.

Crutzen, P.J., A.R. Mosier, K.A. Smith, and W. Winiwarter. 2008. N_2O release from agrobiofuel production negates global warming reduction by replacing fossil fuels. Atmospheric Chemistry and Physics. *Atmospheric Chemistry and Physics* 8:389-395.

Cutler, J.P., 2000. *Iowa – Portrait of the Land.* Iowa Department of Natural Resources, Iowa City, IA

D'Elia, C. F., J.G. Sanders, and W.R. Boynton. 1986. Nutrient enrichment studies in a coastal plain estuary: phytoplankton growth in large scale, continuous cultures. *Canadian Journal of Fisheries and Aquatic Sciences* 43:397-406.

D'Elia, C.F. 1987. Nutrient enrichment of the Chesapeake Bay: too much of a good thing. *Environment* 29:6-11.

David, M.B., L.E. Gentry. 2000. Anthropogenic Inputs of Nitrogen and Phosphorus and Riverine Export for Illinois, USA. *Journal of Environmental Quality* 29(2): 494-508.

David, M.B., G.F. McIsaac, R.G. Darmody, and R.A. Omonde. 2009. Long-term changes in mollisol organic carbon and nitrogen. *Journal of Environmental Quality* 38:200-211.

Day, J.W., W.J. Mitsch, R.R. Lane, and L. Zhang. 2005. *Restoration of Wetlands and Water Quality in the Mississippi-Ohio-Missouri (MOM) River Basin and Louisiana Delta.* Final Report to Louisiana Department of Natural Resources, Baton Rouge, LA, Olentangy River Wetland Research park, The Ohio State University and School of Coast and Environment, Lousiana State University.

Del Grosso, S.J., A.R. Mosier, W.J. Parton, and D.S. Ojima. 2005. DAYCENT model analysis of past and contemporary soil N_2O and net greenhouse gas flux for major crops in the USA. *Soil Tillage and Research* 83:9-24.

Del Grosso, S.J., W.J. Parton, D.S. Ojima, and A.R. Mosier. 2006. Using ecosystem models to inventory and mitigate environmental impacts of agriculture 2006. In *Proceedings Workshop on Agricultural Air Quality: State of the Science,* eds. V.P Aneja, W.H. Schlesinger, R. Knighton, G. Jennings, D. Niyogi, W. Gilliam, and C.S. Duke, 571-574. North Carolina State University, Department of Communication Services, Raleigh, NC, June 5-8, 2006.

deNevers, N. 1995. *Air Pollution Control Engineering,* McGraw Hill, New York.

Dennis, R. 1997. Using the Regional Acid Deposition Model to determine the nitrogen deposition airshed of the Chesapeake Bay watershed. In *Atmospheric Deposition of Contaminants to the Great Lakes and Coastal Waters,* ed. J.E. Baker, 393-413. SETAC Press, Pensacola, FL.

Dentener, F., J. Drevet, J. F. Lamarque, I. Bey, B. Eickhout, A. M. Fiore, D. Hauglustaine, L. W. Horowitz, M. Krol, U. C. Kulshrestha, M. Lawrence, C. Galy-Lacaux, S. Rast, D. Shindell, D. Stevenson, T. Van Noije, C. Atherton, N. Bell, D. Bergman, T. Butler, J. Cofala, B. Collins, R. Doherty, K. Ellingsen, J. Galloway, M. Gauss, V. Montanaro, J. F. Muller, G. Pitari, J. Rodriguez, M. Sanderson, F. Solmon, S. Strahan, M. Schultz, K. Sudo, S. Szopa, and O. Wild. 2006. Nitrogen and sulfur deposition on regional and global scales: A multimodel evaluation. *Global Biogeochemical Cycles* 20(4) 21p.

Department of Sustainability and Environment. 2008. *Bush Tender: Rethinking Investment for Native Vegetation Outcomes. The Application of Auctions for Securing Private Land Management Agreements.* State of Victoria, Department of Sustainability and Environment, East Melbourne, Australia.

DeSimone, L.A., P.A. Hamilton, and R.J. Gilliom. 2009. *Quality of Water from Domestic Wells in Principal Aquifers of the United States, 1991 – 2004. Overview of Major Findings.* U.S. Geological Survey Circular 1332. 48 p.

Diaz, R.J., and R. Rosenberg, R. 1995. Marine benthic hypoxia: a review of its ecological effects and the behavioral responses of benthic macrofauna. *Oceanography and Marine Biology Annual Review* 33:245-303.

Dickerson, R.R., B.G. Doddridge, P. Kelley, and K.P. Rhoads.1995. Large-scale pollution of the atmosphere over the North Atlantic Ocean: Evidence from Bermuda. *Journal of Geophysical Research* 100:8945-8952.

Dobermann, A., and K.G. Cassman. 2004. Environmental dimensions of fertilizer N: what can be done to increase nitrogen use efficiency and ensure global food security? In *Agriculture and the Nitrogen Cycle: Assessing the Impacts of Fertilizer Use on Food Production and the Environment,* eds. A. Mosier and K. Syers, 261-278. SCOPE 65. Island Press, Washington, D.C.

Dobermann, A., and K.G. Cassman. 2005. Cereal area and nitrogen use efficiency are drivers of future nitrogen fertilizer consumption. *Science in China Series C. Life Sciences,* 48:745-758.

Doddridge, B.G., R.R. Dickerson, J.Z. Holland, J.N. Cooper, R.G. Wardell, O. Poulida, and J.G. Watkins 1991. Observations of tropospheric trace gases and meteorology in rural Virginia using an unattended monitoring-system-Hurricane Hugo (1989), a Case-Study. *Journal of Geophysical Research-Atmospheres* 96: 9341-9360.

Doddridge, B.G., R.R. Dickerson, R.G. Wardell, K.L. Civerolo, and L.J. Nunnermacker. 1992. Trace gas concentrations and meteorology in rural Virginia. 2. Reactive nitrogen-compounds. *Journal of Geophysical Research-Atmospheres* 97: 20631-20646

Dodds, W.K., K.R. Johnson, and J.C. Priscu. 1989. Simultaneous nitrogen and phosphorus deficiency in natural phytoplankton assemblages: theory, empirical evidence and implications for lake management. *Lake and Reservoir Management* 5:21-26.

Doering, O.C., F. Diaz-Hermelo, C. Howard, R. Heimlich, F. Hitzhusen, R. Kazmierczak, J. Lee, L. Libby, W. Milon, T. Prato, and M. Ribaudo. 1999. *Evaluation of the Economic Costs and Benefits of Methods for Reducing Nutrient Loads to the Gulf of Mexico.* Topic 6 Report for the Integrated Assessment on Hypoxia in the Gulf of Mexico. U.S. Department of Commerce, NOAA, Washington D.C.

Doering, O., and W. Tyner. 2009. U.S. and international policies affecting liquid biofuels' expansion and profitability. *International Journal of Biotechnology,* 11(1 and 2): 150-167.

Donahue, N.N., A.L. Robinson, and S.N. Pandis 2009. Atmospheric organic particulate matter: From smoke to secondary organic aerosol. *Atmospheric Environment* 43:97-109.

Doney, S.C., N. Mahowald, I. Lima, R. Feely, F.T. Mackenzie, J.F. Lamarque, and P.J. Rasch. 2007. Impact of anthropogenic atmospheric nitrogen and sulfur deposition on ocean acidification and the inorganic carbon system. *Proceedings of the National Academy of Sciences of the United States of America* 104:14580-14585.

Dortch, Q. 1990. The interaction between ammonium and nitrate uptake in phytoplankton. *Marine Ecology Progress Series* 61:183-201.

Dortch, Q., and T.E. Whitledge. 1992. Does nitrogen or silicon limit phytoplankton production in the Mississippi River plume and nearby regions? *Continental Shelf Research* 12:1293-1309.

Duce, R.A., K. LaRoche, K.R. Altieri, K.R. Arrigo, A.R. Baker, D.G. Capone, S. Cornell, F. Dentener, J. Galloway, R.S. Ganeshram, R.J. Geider, T. Jickells, M.M. Kuypers, R. Langlois, P.S. Liss, S.M. Liu, J.J. Middelburg, C.M. Moore, S. Nickovic, A. Oschlies, T. Pedersen, J. Prospero, R. Schlitzer, S. Seitzinger, L.L. Sorensen, M. Uematsu, O. Ulloa, M. Voss, B. Ward, and L. Zamora. 2008. Impacts of atmospheric anthropogenic nitrogen on the open ocean. *Science* 320:893-897.

Dugdale, R.C. 1967. Nutrient limitation in the sea: Dynamics, identification and significance. *Limnology and Oceanography* 12:685-695.

Dunlea, E.J., S.C. Herndon, D.D. Nelson, R.M. Volkamer, F. San Martini, P.M. Sheehy, M.S. Zahniser, J.H. Shorter, J.C. Wormhoudt, B.K. Lamb, E.J. Allwine, J.S. Gaffney, N.A. Marley, M. Grutter, C. Marquez, S. Blanco, B. Cardenas, A. Retama,C.R R. Villegas, C.E. Kolb, L.T. Molina, and M.J. Molina. 2007. Evaluation of nitrogen dioxide chemiluminescence monitors in a polluted urban environment. *Atmospheric Chemistry and Physics* 7: 2691-2704.

Duvick, D.N., and K.G. Cassman. 1999. Post-Green revolution trends in yield potential of temperate maize in the North-Central United States. *Crop Science* 39:1622-1630.

Ecology and Oceanography of Harmful Algal Blooms (ECOHAB). 1995. *The Ecology and Oceanography of Harmful Algal Blooms: A National Research Agenda.* Woods Hole Oceanographic Institution, Woods Hole, MA

Elmgren, R., and U. Larsson. 2001. Nitrogen and the Baltic Sea: Managing nitrogen in relation to phosphorus. *The Scientific World* 1(S2), 371-377.

Elser, J.J., E.R. Marzolf, and C.R. Goldman. 1990. Phosphorus and nitrogen limitation of phytoplankton growth in the freshwaters of North America: a review and critique of experimental enrichments. *Canadian Journal of Fisheries and Aquatic Sciences* 47: 1468-1477.

Elser, J.J., M.E.S. Bracken, E.E. Cleland, D.S. Gruner, W.S. Harpole, H. Hillebrand, J.T. Bgai, E.W. Seabloom, J.B. Shurin, and J.E. Smith. 2007. Global analysis of nitrogen and phosphorus limitation of primary producers in freshwater, marine and terrestrial ecosystems. *Ecology Letters* 10:1124-1134.

Elser, J.J., T. Andersen, J.S. Baron, A.-K. Bergstrom, M. Jansson, M. Kyle, K.R. Nydick, S.L. Steger, and D.O. Hessen. 2009. Shifts in lake N:P stoichiometry and nutrient limitation driven by atmospheric nitrogen deposition. *Science* 326:835-837.

Emmons, L.K., M.A. Carroll, D.A. Hauglustaine, G.P. Brasseur, C. Atherton, J. Penner, S. Sillman, H. Levy, F. Rohrer, W.M.F. Wauben, P.F.J. Van Velthoven, Y. Wang, D. Jacob, P. Bakwin, R. Dickerson, B. Doddridge, C. Gerbig, R. Honrath, G. Hubler, D. Jaffe, Y. Kondo, J.W. Munger, A. Torres, and A. Volz Thomas. 1997. Climatologies of NO_x and NOy: A comparison of data and models. *Atmospheric Environment* 31:1851-1904.

Erisman, J., M.A. Sutton, and J. N. Galloway. 2008. How a century of ammonia synthesis changed the world. *Nature Geoscience* 1:636-639

Ezzati M., A.D. Lopez, A. Rodgers, and C.J.L. Murray. 2004. *Comparative Quantification of Health Risks: Global and Regional Burden of Disease Attributable to Selected Major Risk Factors.* World Health Organization, Geneva.

Fahey, D.W., G. Huebler, D.D. Parrish, E.J. Williams, R.B. Norton, B A. Ridley, H.B. Singh, S.C. Liu, and F.C. Fehsenfeld. 1986. Reactive nitrogen species in the troposphere: Measurements of NO, NO_2, HNO_3, particulate nitrate, pereoxyacetyl nitrate (PAN), O_3, and total reactive odd-nitrogen (NO_y) at Niwot Ridge, Colorado. *Journal of Geophysical Research* 91:9781-9793.

FAO (Food and Agricultural Organization of the United Nations). 2007. *FAO Database Collections.* www.apps.fao.org. Rome, Italy: FAO

FAO (Food and Agriculture Organization of the United Nations). 2010a. *FAOSTAT.* http://faostat.fao.org/ FAOSTAT Agricultural Data Available on the World Wide Web

FAO (Food and Agriculture Organization of the United Nations). 2010b. *Major Food and Agricultural Commodities Producers.* www.fao.org/es/ess/top/country.html

Fehsenfeld, F.C., R.R. Dickerson, G. Huebler, W.T. Luke, L. Nunnermacker, E.J. Williams, J. Roberts, J.G. Calvert, C. Curran, A.C. Delany, C.S. Eubank, D W. Fahey, A. Fried, B. Gandrud, A. Langford, P. Murphy, R.B. Norton, K. Pickering, and B. Ridley. 1987. A ground-based intercomparison of NO, NO_x, NO_y measurement techniques. *Journal of Geophysical Research* 92:14710-14722

Fenn, M.E., R. Haeuber, G.S. Tonnesen, J.S. Baron, S. Grossman-Clarke, D. Hope, D.A. Jaffe, S. Copeland, L. Geiser, H.M. Rueth, and J.O. Sickman. 2003. Nitrogen Emissions, Deposition, and Monitoring in the Western United States. *BioScience* 53:391-403.

Ferber, L.R., S.N. Levine, A. Lini, and G.P. Livingston. 2004. Do cyanobacteria dominate in eutrophic lakes because they fix atmospheric nitrogen? *Freshwater Biology* 49:690-708.

Fehsenfeld, F.C., R.R. Dickerson, G. Huebler, W.T. Luke, L. Nunnermacker, E.J. Williams, J. Roberts, J.G. Calvert, C. Curran, A.C. Delany, C.S. Eubank, D.W. Fahey, A. Fried, B. Gandrud, A. Langford, P. Murphy, R.B. Norton, K. Pickering, and B. Ridley. 1987. A ground-based intercomparison of NO, NO_x, NO_y measurement techniques. *Journal of Geophysical Research* 92:14710-14722.

Fishman, J., and P.J. Crutzen.1978. The origin of ozone in the troposphere. *Nature* 274:855.

Fishman, J., S. Solomon, and P.J. Crutzen.1979. Observational and theoretical evidence in support of a significant in situ photochemical source of tropospheric ozone. *Tellus* 31: 432-446.

Fixen, P.E. 2005. Decision Support Systems. In *Integrated Crop Nutrient Management, International Fertilizer Society Proceedings,* 1-31.

Fixen, P.E. 2005. Understanding and improving nutrient use efficiency as an application of information technology. In *Proceedings of the Symposium on Information Technology in Soil Fertility and Fertilizer Management, a satellite symposium at the XV International Plant Nutrient Colloquim, Sep. 14–16, 2005. Beijing, China.*

Fixen, P.E., and F.B. West. 2002. Nitrogen Fertilizers: Meeting Contemporary Challenges. *AMBIO: A Journal of the Human Environment* 31(2):169-176.

Flores, E., and A. Herrero. 2005. Nitrogen assimilation and nitrogen control in cyanobacteria. *Biochemical Society Transactions* 33(1):164-167.

Forbes, M.G., R.D. Doyle, J.T. Scott, J.K. Stanley, H. Huang, and B.W. Brooks. 2008. Physical factors control phytoplankton production and nitrogen fixation in eight Texas reservoirs. *Ecosystems* 11:1181-1197.

Freney, J.R., J.R. Simpson, and O.T. Denmead. 1983. Volatilization of ammonia. In *Gaseous Loss of Nitrogen from Plant-Soil Systems,* eds. J.R. Freney and J.R. Simpson, 1-32. Kluwer Academic Publishers, The Hague.

Funk, T.L., R. Hussey, Y. Zhang, and M. Ellis. 2004a. Synthetic covers for emissions control from earthen embanked swine lagoons - Part I: Positive pressure lagoon cover. *Applied Engineering in Agriculture* 20:233-238.

Funk, T.L., A. Mutlu, Y. Zhang, and M. Ellis. 2004b. Synthetic covers for emissions control from earthen embanked swine lagoons - part II: Negative pressure lagoon cover. *Applied Engineering in Agriculture* 20:239-242.

Galloway, J.N., and E.B. Cowling. 2002. Reactive nitrogen and the world: 200 years of change. *AMBIO: A Journal of the Human Environment* 31:64-71.

Galbally, I.E., and C.R. Roy. 1978. Loss of fixed nitrogen from soils by nitric oxide exhalation. *Nature* 275:734-735.

Galloway, J.N., and D.M. Whelpdale. 1987. WATOX-86 overview and western North Atlantic Ocean and N atmospheric budgets. *Global Biogeochemical Cycles* 1:261-281.

Galloway, J.N., D.M. Whelpdale, and G.T. Wolff. 1984. The Flux of S and N Eastward from North-America. *Atmospheric Environment* 18:2595-2607.

Galloway, J.N., R.S. Artz, U. Dayan, R.F. Pueschel, and J. Boatman. 1988. WATOX-85 An aircraft and ground sampling program to determine the transport of trace gases and aerosols across the western Atlantic Ocean. *Atmospheric Environment* 22:2345-2360.

Galloway, J.N., W.H. Schlesinger, H. Levy II, A. Michaels, and J.L. Schnoor. 1995. Nitrogen fixation: Anthropogenic enhancement-environmental response. *Global Biogeochemical Cycles* 9:235-252.

Galloway, J.N., J.D. Aber, J.W. Erisman, S.P. Seitzinger, R.H. Howarth, E.B. Cowling, and B.J. Cosby. 2003. The nitrogen cascade. *Bioscience* 53:341-356.

Galloway, J.N., F.J. Dentener, D.G. Capone, E.W. Boyer, R.W. Howarth, S.P. Seitzinger, G.P. Asner, C.C. Cleveland, P.A. Green, E.A. Holland, D.M. Karl, A.F. Michaels, J.H. Porter, A.R. Townsend, and C.J. Vorosmarty. 2004. Nitrogen cycles: past, present, and future. *Biogeochemistry* 70:153-226.

Galloway, J.N., M. Burke, G.E. Bradford, R. Naylor, W. Falcon, A.K. Chapagain, J.C. Gaskell, E. McCullough, H.A. Mooney, K.L.L. Oleson, H. Steinfeld, T. Wassenaar, and V. Smil. 2007. International trade in meat: The tip of the pork chop. *AMBIO: A Journal of the Human Environment* 36(8):622-629.

Galloway, J.N., A.R. Townsend, J.W. Erisman, M.Bekunda, Z. Cai, J.R. Freney, L.A. Martinelli, S.P. Seitzinger, and M.A. Sutton. 2008. Transformation of the nitrogen cycle: recent trends, questions and potential solutions. *Science* 320:889-892.

Gardner, W.S., P.J. Lavrentyev, J.F. Cavaletto, M.J. McCarthy, B.J. Eadie, T.H. Johengen, and J.B. Cotner. 2004. The distribution and dynamics of nitrogen and microbial plankton in southern Lake Michigan during spring transition 1999-2000. *Journal of Geophysical Research* 109:CO3007, doi:10.1029/2002JC001588, 1-16.

Garner, J.H.B., T. Pagano and E.B. Cowling. 1989. *An Evaluation of the Role of Ozone, Acid Deposition, and Other Airborne Pollutants in the Forest of Eastern North America.* Gen. Tech. Rep. SE-59, U.S. Dept. of Agriculture, Forest Service, S.E. Forest Experiments Station, Asheville, North Carolina

Gego, E., S.P. Porter, A. Gilliland, and S. Trivikrama Rao. 2007. Observation-based assessment of the impact of nitrogen oxides emissions reductions on ozone air quality over the eastern United States. *Journal of Applied Meteorology and Climatology* 46:994-1008.

Geosyntec Consultants, Wright Water Engineers, Inc. 2008. *Analysis of Treatment System Performance. International Stormwater Best Management Practices (BMP) Database (1999-2008).* Water Environment Research Foundation et al., Alexandria, VA. 20 p.

Gilbert, P.M., J. Harrison, C. Heil, and S. Seitzinger. 2006. Escalating worldwide use of urea - a global change contributing to coastal eutrophication. *Biogeochemistry* 77:441-463.

Giller, K.E., P. Chalk, A. Dobermann, L. Hammond, P. Heffner, J.K. Ladha, P. Nyamudeza, L. Maene, H. Ssali, and J. Freney. 2004. Emerging Technologies to Increase the Efficiency of use of Fertilizer Nitrogen. In *Agriculture and the Nitrogen Cycle,* eds. A.R. Mosier, J. K. Syers and J.R. Freney, 35-51. SCOPE 65, Island Press, Washington D.C.

Gilliland, A.B., R.L. Dennis, S.J. Roselle, and T.E. Pierce. 2003. Seasonal NH_3 emission estimates for the eastern United States based on ammonium wet concentrations and an inverse modeling method. *Journal of Geophysical Research-Atmospheres* 108(D15): 4477, doi:10.1029/2002JD003063.

Gilliland, A.B., K.W. Appel, R.W. Pinder, and R.L. Dennis. 2006. Seasonal NH_3 emissions for the continental united states: Inverse model estimation and evaluation. *Atmospheric Environment* 40:4986-4998.

Gilliland, A.B., C. Hogrefe, R.W. Pinder, J.M. Godowitch, K.L. Foley, and S.T. Rao. 2008. Dynamic evaluation of regional air quality models: Assessing changes in O_3 stemming from changes in emissions and meteorology. *Atmospheric Environment* 42:5110-5123.

Goldenberg, S.B., C.W. Landsea, A.M. Mestas-Nues, and W.M. Gray. 2001. The recent increase in Atlantic Hurricane Activity: Causes and implications. *Science* 293:474-479.

Goldman, C.R. 1981. Lake Tahoe: Two decades of change in a nitrogen deficient oligotrophic lake. *Verhandlungen der Internationalen Vereinigung für Theoretische und Angewandte Limnologie* 2:45-70.

Goldman, C.R. 1988. Primary productivity, nutrients, and transparency during the early onset of eutrophication in ultra-oligotrophic Lake Tahoe, California-Nevada. *Limnology and Oceanography* 33:1321-1333.

Goldman, C.R. 2002. Lessons in critical ecosystem protection: the role of science in management decisions at Lake Tahoe CA/NV. In *Summary Report, Workshop on Critical Ecosystem Assessment, U.S. EPA, June 17–20, 2002, Keystone, CO,* U.S. EPA Office of Science Policy (OSP), Office of Research and Development (ORD), Washington, DC. [Graphics accompanying the report are available at: http://epa.gov/osp/presentations/critical/goldman.pdf.]

Goulding, K. 2004. Pathways and losses of fertilizer nitrogen at different scales. In *Agriculture and the Nitrogen Cycle,* eds. A.R. Mosier, J.K. Syers and J.R. Freney, 209-219. SCOPE 65. Island Press, Washington, DC.

Greenwood, D.J., G. Lemaire, G. Gosse, P. Cruz, A. Draycott, A., and J.T. Neeteson. 1990. Decline in percentage N of C3 and C4 crops with increasing plant mass. *Annals of Botany* 66:425-436.

Guillard, K. 2008. *New England Regional Nitrogen and Phosphorus Fertilizer and Associated Management Practice Recommendations for Lawns Based on Water Quality Considerations.* University of Maine Cooperative Extension, Orono, ME

Hall, K.D., J. Guo, M. Dore, and C.C. Chow. 2009. The progressive increase of food waste in America and its environmental impact. *PLoS One* 4(11):e7940. doi:10.1371/journal.pone.0007940

Hargrove, J., and J.S. Zhang. 2008. Measurements of NO_x, acyl peroxynitrates, and NO_y with automatic interference corrections using a NO_2 analyzer and gas phase titration. *Review of Scientific Instruments* 79(4):046109.

Harper, L.A., R.R. Sharpe, and T.B. Parkin. 2000. Gaseous nitrogen emissions from anaerobic swine lagoons: Ammonia, nitrous oxide, and dinitrogen gas. *Journal of Environmental Quality* 29:1356-1365.

Harrington, M.B. 1999. *Responses of Natural Phytoplankton Communities from the Neuse River Estuary, NC to Changes in Nitrogen Supply and Incident Irradiance.* MSc. thesis, University of North Carolina, Chapel Hill, North Carolina. 89p.

Harrison, P., and D. Turpin. 1982. The manipulation of physical, chemical, and biological factors to select species from natural phytoplankton populations. In *Marine Mesocosms: Biological and Chemical Research in Experimental Ecosystems,* eds. G. Grice and M. Reeve, 275-287. Springer-Verlag, New York.

Havens, K.E., K.R. Jin, N. Iricanin, and R.T. James. 2007. Phosphorus dynamics at multiple time scales in the pelagic zone of a large shallow lake in Florida, USA. *Hydrobiologia* 581:25-42.

Havenstein, G. B., P.R. Ferket, and S.E. Scheideler. 1994. Carcass composition and yield of 1991 vs.1957 broilers when fed typical 1957 vs. 1991 broiler diets. *Poultry Science* 73:1785-1804.

Heck, W.W., W.W. Cure, J.O. Rawlings, L.J. Zargoza, A.S. Heagle, H.E. Heggestad, R.J. Kohut, L.W. Kress, and P.J. Temple. 1984. Assessing impacts of ozone on agricultural crops. I. Overview. *Journal of the Air Pollution Control Association* 34:729-735.

Hettelingh, J-P., M. Posch, and P.A.M. De Smet. 2001. Multi-Effect Critical Loads Used in Multi-Pollutant Reduction Agreements in Europe. *Water, Air, and Soil Pollution* 130:1133-1138.

Hernandez, M.E., and W.J. Mitsch. 2007. Denitrification in created riverine wetlands: Influence of hydrology and season. *Ecological Engineering* 30:78-88.

Hey, D.L. 2002. Nitrogen Farming: Harvesting a Different Crop. *Restoration Ecology* 10(1):1-10.

Hey, D.L., and N.P. Philippi. 1995. Flood reduction through wetland restoration: the upper Mississippi River basin as a case history. *Restoration Ecology* 3 (1):4-17.

Hey, D.L., D.L. Montgomery, L.S. Urban, T. Prato, F. Andrew, F., M. Martel, J. Pollack, Y. Steele, and R. Zarwell. 2004. *Flood Damage Reduction in the Upper Mississippi River Basin: An Ecological Alternative.* The McKnight Foundation, Minneapolis, MN

Hey, D.L., J.A. Kostel, A.P. Hurter, and R.H. Kadlec. 2005a. *Comparing Economics of Nitrogen Farming with Traditional Removal.* WERF 03-WSM-6CO. Water and Environment Research Foundation, Alexandria, VA.

Hey, D.L., L.S. Urban, and J.A. Kostel. 2005b. Nutrient farming: The business of environmental management. *Ecological Engineering* 24:279-287.

Hicks, B.B. 2006. Dry deposition to forests - On the use of data from clearings. *Agricultural and Forest Meteorology* 136:214-221.

Hicks, B.B., R.P. Hosker Jr., T.P. Meyers, and J.D. Womack. 1991. Dry Deposition Inferential Measurement Techniques.1. Design and Tests of a Prototype Meteorological and Chemical-System for Determining Dry Deposition. *Atmospheric Environment Part A. General Topics* 25:2345-2359.

Holland, E.A., and J.F. Lamarque. 1997. Modeling bio-atmospheric coupling of the nitrogen cycle through NO emissions and NOy deposition. *Nutrient Cycling in Agroecosystems* 48:7-24.

Holland, E.A., B.H. Braswell, J.F. Lamarque, A. Townsend, J. Sulzman, J.F. Muller, F. Dentener, G. Brasseur, H. Levy, J.E. Penner, and G.J. Roelofs. 1997. Variations in the predicted spatial distribution of atmospheric nitrogen deposition and their impact on carbon uptake by terrestrial ecosystems. *Journal of Geophysical Research-Atmospheres* 102:15849-15866.

Holland, E.A., B.H. Braswell, J. Sulzman, and J.F. Lamarque. 2005. Nitrogen deposition onto the United States and Western Europe: Synthesis of observations and models. *Ecological Applications* 15:38-57.

Holland, E., F. Dentener, B. Braswell, and J. Sulzman. 1999. Contemporary and preindustrial global reactive nitrogen budgets. *Biogeochemistry* 43:7-43.

Horii, C.V., J.W. Munger, S.C. Wofsy, M. Zahniser, D. Nelson, and J.B. McManus. 2004. Fluxes of nitrogen oxides over a temperate deciduous forest. *Journal of Geophysical Research-Atmospheres,* 109, D08305, doi:10.1029/2003JD004326.

Horii, C.V., J.W. Munger, S.C. Wofsy, M. Zahniser, D. Nelson, and J.B. McManus. 2006. Atmospheric reactive nitrogen concentration and flux budgets at a Northeastern U.S. forest site. *Agricultural and Forest Meteorology* 136:159-174.

Horowitz, L.W., J. Liang, G.M. Gardner, and D.J. Jacob. 1998. Export of reactive nitrogen from North America during summertime: Sensitivity to hydrocarbon chemistry. *Journal of Geophysical Research Atmospheres* 103:13451-13476.

Horowitz, L.W., A.M. Fiore, G.P. Milly, R.C. Cohen, A.Perring, P. J. Wooldridge, P.G. Hess, L. K. Emmons, and J. Lamarque. 2007. Observational constraints on the chemistry of isoprene nitrates over the eastern United States. *Journal of Geophysical Research-Atmospheres* 112, D12S08, doi:10.1029/2006JD007747

Hov, Ø., B.A. Hjøllo, and A. Eliassen. 1994. Transport distance of ammonia and ammonium in Northern Europe 1. Model description. *Journal of Geophysical Research* 99:18735-18748.

Howarth, R.W. 1998. An assessment of human influences on inputs of nitrogen to the estuaries and continental shelves of the North Atlantic Ocean. *Nutrient Cycling in Agroecosystems* 52:213-223.

Howarth, R.W., G. Billen, D. Swaney, A. Townsend, N. Jaworski, K. Lajtha, J.A. Downing, R. Elmgren, N. Caraco, T. Jordan, F. Berendse, J. Freney, V. Kudeyarov, P. Murdoch, and Z. Zhao-Liang. 1996. Regional nitrogen budgets and riverine N & P fluxes for the drainages to the North Atlantic Ocean: Natural and human influences. *Biogeochemistry* 35:75-139.

Howarth, R.W., E.W. Boyer, W.J. Pabich, and J.N. Galloway. 2002. Nitrogen use in the United States from 1961-2000 and potential future trends. *AMBIO: A Journal of the Human Environment* 31:88-96.

Hu, X., G.F. McIsaac, M.B. David, and C.A.L. Louwers. 2007. Modeling riverine nitrate export from an East-Central Illinois watershed using SWAT. *Journal of Environmental Quality* 36:996-1005.

Hudman, R.C., D.J. Jacob, S. Turquety, M. Leibensperger, L.T. Murray, S. Wu, A.B. Gilliland, M. Avery, T.H. Bertram, W. Brune, R.C. Cohen, J.E. Dibb, F.M. Flocke, A. Fried, J. Holloway, J.A. Neuman, R. Orville, A. Perring, X. Ren, W. Sachse, H.B. Singh, A. Swanson, and P.J. Wooldridge. 2007. Surface and lightning sources of nitrogen oxides over the United States: Magnitudes, chemical evolution, and outflow. *Journal of Geophysical Research-Atmospheres* 112, D12S05, doi:10.1029/2006JD007912 .

Hungate, B.A., J.S. Dukes, M.R. Shaw, Y.Q. Luo, and C.B. Field. 2003. Nitrogen and climate change. *Science* 302:1512-1513.

Hungate, B.A., P.D. Stiling, P. Dijkstra, D.W. Johnson, M.E. Ketterer, G.J. Hymus, C.R. Hinkle, and B.G. Drake. 2004. CO_2 elicits long-term decline in nitrogen fixation. *Science* 304:1291-1291.

Husar, R.B., D.E. Patterson, C.C. Paley, and N.G. Gillani. 1977. Ozone in hazy air masses. In *Proceedings of the International Conference on Photochemical Oxidant Pollution and Its Control, September, 1976,* 275–282. EPA-600/3-77-001, U.S. EPA, Raleigh, NC

IFA. 2004. *International Fertilizer Industry Association. Current Situation and Prospects for Fertilizer Use in Sub-Saharan Africa,* a paper presented by Luc Maene at the symposium Fertilizer Nitrogen and Crop Production in Africa, in Kampala, Uganda, January 14, 2004.

IPCC. 2001. *Intergovernmental Panel on Climate Change. Technical Summary of the 3rd Assessment Report of Working Group 1.* D.L. Albritton and L.G. Meira Filho (Coordinating lead authors). World Meteorological Organisation (WMO) and the United Nations Environment Programme (UNEP), 63 p.

IPCC. 2007a. *Intergovernmental Panel on Climate Change: Fourth Assessment Report: Climate Change 2007* (AR4). www.ipcc.ch/publications_and_data/publications_and_data_reports.htm#1 [accessed September 10, 2010]

IPCC. 2007b. *Working Group III Contribution to the Intergovernmental Panel on Climate Change Fourth Assessment Report Climate Change 2007: Mitigation of Climate Change,* Cambridge University Press, Cambridge, UK.

James, K.M. 2008. *The Development of U.S. Ammonia Emission Factors for use in Process Based Modeling.* M.S. Thesis, North Carolina State University, Raleigh, NC.

Jaworski, N., R. Howarth, and L. Hetling. 1997. Atmospheric deposition of nitrogen oxides onto the landscape contributes to coastal eutrophication in the Northeast United States. *Environmental Science and Technology* 31:1995-2004.

Jaynes, D.B., and Karlen, D.L. 2005. Sustaining soil resources while managing nutrients. In *Proceedings of the Upper Mississippi River-Sub Basin Hypoxia Nutrient Committee Workshop, Aimes, Iowa, September 26-28, 2005,*141-150. www.umrshnc.org/files/hypwebversion.pdf [accessed September 10, 2010]

Jha, M., C.F. Wolter, K.E. Schilling, and P.W. Gassman. 2010. TMDL analysis with SWAT modeling for the Raccoon River watershed in Iowa. *Journal of Environmental Quality* 39:1317-1327.

Johnson, A.H., and T.G. Siccama. 1983. Acid Deposition and Forest Decline. *Environmental Science and Technology* 17:294a-305a.

Justić, D., N.N. Rabalais, and R.E. Turner. 1995a. Stoichiometric nutrient balance and origin of coastal eutrophication. *Marine Pollution Bulletin* 30:41-46.

Justić, D., N.N. Rabalais, R.E. Turner, R.E., and Q. Dortch. 1995b. Changes in nutrient structure of river-dominated coastal waters: Stoichiometeric nutrient balance and its consequences. *Estuarine, Coastal, and Shelf Science* 40: 339-356.

Kadlec, R.H., and S.D. Wallace. 2009. *Treatment Wetlands,* 2nd ed. CRC Press, Boca Raton, FL.

Kalkhoff, S.J., K.K. Barnes, K.D. Becher, M.E. Savoca, D.J. Schnoebelen, E.M. Sadorf, S.D. Porter, and D.J. Sullivan. 2000. *Water Quality in the Eastern Iowa Basins, Iowa and Minnesota, 1996–98.* U.S. Geological Survey Circular 1210, 37p. [available at: http://pubs.water.usgs.gov/circ1210/]

Kantor, L.S., K. Lipton, A. Manchester, and V.Oliveira. 1997. Estimating and addressing America's food losses. *Food Review* 20:2-12.

Kashihira, N., K. Makino, K. Kirita, and Y. Watanabe. 1982. Chemi-luminescent nitrogen detector gas-chromatography and its application to measurement of atmospheric ammonia and amines. *Journal of Chromatography* 239:617-624.

Kasibhatla, P.S., H. Levy II, and W.J. Moxim. 1993. Global NO_x, HNO_3, PAN, and NO_y distributions from fossil-fuel combustion emissions - a model study. *Journal of Geophysical Research-Atmospheres* 98:7165-7180.

Keene, W.C., J.A. Montag, J.R. Maben, M. Southwell, J. Leonard, T.M. Church, J.L. Moody, and J.N. Galloway. 2002. Organic nitrogen in precipitation over Eastern North America. *Atmospheric Environment* 36:4529-4540.

Kelly, V.R., G.M. Lovett, K.C. Weathers, and G.E. Likens. 2002. Trends in atmospheric concentration and deposition compared to regional and local pollutant emissions at a rural site in southeastern New York, USA. *Atmospheric Environment* 36:1569-1575.

Kermarrec, C., P. Robin, N. Bernet, F. Trolard, P.Armando de Oliveira, A. Laplanche, and D. Souloumiac. 1998. Influence du mode de ventilation des litières sur les émissions gazeuses d'azote NH_3, N_2O, N_2 et sur le bilan d'azote en engraissement porcin. *Agronomie* 18:473-488.

Kim, D-S., V.P. Aneja, and W.P. Robarge. 1994. Characterization of nitrogen oxide fluxes from soil of a fallow field in the central Piedmont of North Carolina. *Atmospheric Environment* 28:1129-1137.

Kleinman, L.I., P.H. Daum, Y.N. Lee, G.I. Senum, S.R. Springston, J. Wang, C. Berkowitz, J. Hubbe, R.A. Zaveri, F.J. Brechtel, J. Jayne, T.B. Onasch, and D. Worsnop. 2007. Aircraft observations of aerosol composition and ageing in New England and Mid-Atlantic States during the summer 2002 New England Air Quality Study field campaign. *Journal of Geophysical Research-Atmospheres* 112, D09310, doi:10.1029/2006JD007786.

Klopfenstein, T.J., G.E. Erickson, and V.R. Bremer. 2008. Board-Invited Review: Use of Distillers Byproducts in the Beef Cattle Feeding Industry. *Journal of Animal Science* 86:1223-1231.

Knobeloch, L., and M. Proctor 2001. Eight Blue Babies. *Wisconsin Medical Journal* 100 (8): 43-47.

Koebel, M., M. Elsener, O. Krocher, C. Schar, R. Rothlisberger, F. Jaussi, and M. Mangold. 2004. NO_x reduction in the exhaust of mobile heavy-duty diesel engines by urea-SCR. *Topics in Catalysis* 30-31(1):43-48.

Kohn, R.A. 2004. Use of animal nutrition to manage nitrogen emissions from animal agriculture. In *Mid-Atlantic Nutrition Conference,* 25-30. University of Maryland, College Park, MD.

Kohn, R.A., Z. Dou, J.D. Ferguson, and R.C. Boston. 1997. A sensitivity analysis of nitrogen losses from dairy farms. *Journal of Environmental Management* 50:417-428.

Kratzer, C.R., and P.L. Brezonik. 1981. A Carlson-type trophic state index for nitrogen in Florida lakes. J*ournal of the American Water Resources Association* 17(4):713-715.

Kreuter, M.W., C. De Rosa, E.H. Howze, and G.T. Baldwin. 2004. Understanding Wicked Problems: A Key to Advancing Environmental Health Promotion. *Health, Education and Behavior* 31:441-54.

Krupnick, A. V. McConnell, D. Austin, M. Cannon, T. Stoessell, and B. Morton. 1998. *The Chesapeake Bay and Control of NO_x Emissions: A Policy Analysis.* Resources for the Future, Washington, DC.

Ladha, J.K., H. Pathak, T.J. Krupnik, J. Six , and C. van Kessel. 2005. Efficiency of Fertilizer Nitrogen in Cereal Production: Retrospects and Prospects. *Advances in Agronomy* 87: 85-176.

Lal, R., J.M. Kimble, R.F. Follett, and C.V. Cole. 1998. *The Potential of U.S. Cropland to Sequester Carbon and Mitigate the Greenhouse Effect.* Ann Arbor Press, Chelsea, MI, 128 p.

Larson, U.R., R. Elmgren, and F. Wulff. 1985. Eutrophication and the Baltic Sea: Causes and Consequences. *AMBIO: A Journal of the Human Environment.* 14:9-14.

The Lawn Institute. 2007. *Turfgrass Producers International.* www.turfgrasssod.org/lawninstitute.html

Lelieveld, J.P., J. Crutzen,V. Ramanathan, M.O. Andreae, C.A.M. Brenninkmeijer, T. Campos, G.R. Cass, R.R. Dickerson, H. Fischer, J.A. de Gouw, A. Hansel, A. Jefferson, D. Kley, A.T. J. de Laat, S. Lal, M.G. Lawrence, J.M. Lobert, O.L. Mayol-Bracero, A.P. Mitra, T. Novakov, S.J. Oltmans, K.A. Prather, T. Reiner, H. Rodhe, H.A. Scheeren, D. Sikka, and J. Williams. 2001. The Indian Ocean Experiment: Widespread air pollution from South and Southeast Asia. *Science* 291:1031-1036.

Levy, H., M.D. Schwarzkopf, L. Horowitz, V. Ramaswamy, and K.L. Findell. 2008. Strong sensitivity of late 21st century climate to projected changes in short-lived air pollutants. J*ournal of Geophysical Research-Atmospheres* 113, D06102, doi:10.1029/2007JD009176.

Lewis, W.M. Jr. 2002. Causes for the high frequency of nitrogen limitation in tropical lakes. *Verhandlungen des Internationalen Verein Limnologie* 28:210-213.

Lewis, W.M. Jr., and W.A Wurtsbaugh. 2008. Control of Lacustrine Phytoplankton by nutrients: Erosion of the Phosphorus Paradigm. *International Review of Hydrobiology* 93(4-5):446-465.

Li, Q.B., D.J. Jacob, J.W. Munger, R.M. Yantosca, and D.D. Parrish. 2004. Export of NO_y from the North American boundary layer: Reconciling aircraft observations and global model budgets. *Journal of Geophysical Research-Atmospheres* 109 (noD2): D02313.1-D02313.12

Liang, J.Y., L.W. Horowitz, D.J. Jacob, Y.H. Wang, A.M. Fiore, J.A. Logan, G.M. Gardner, and J.W. Munger. 1998. Seasonal budgets of reactive nitrogen species and ozone over the United States, and export fluxes to the global atmosphere. *Journal of Geophysical Research-Atmospheres* 103:13435-13450.

Likens, G.E., D.C. Buso, and T.J. Butler. 2005. Long-term relationships between SO_2 and NO_x emissions and SO_4^{2-} and NO_3^- concentration in bulk deposition at the Hubbard Brook Experimental Forest, NH. *Journal of Environmental Monitoring* 7:964-968.

Libra, M., C.F. Wolter, and R.J. Langel. 2004. *Nitrogen and Phosphorus Budgets for Iowa and Iowa Watersheds.* Iowa Geological Survey Technical Information Series 47, Iowa Department of Natural Resources-Geological Survey, Iowa City, IA.

Logan, J.A. 1989. Ozone in rural areas of the United States. *Journal of Geophysical Research* 94: 8511-8532.

Luke, W.T., and R.R. Dickerson. 1987. The flux of reactive nitrogen compounds from eastern North America to the western Atlantic Ocean. *Global Biogeochemical Cycles* 1:329-343.

Luke, W.T., R.R. Dickerson, W.F. Ryan, K.E. Pickering, and L.J. Nunnermacker. 1992. Tropospheric chemistry over the lower Great Plains of the United States II: Trace gas profiles and distributions. *Journal of Geophysical Research* 97:20747-20670.

Luke, W.T., P. Kelley, B.L. Lefer, and M. Buhr. 2010. Measurements of primary trace gases and NO_y composition in Houston, Texas. *Atmospheric Environment* 44(33):4068-4080.

Luria, M., R. J. Valente, S. Bairai, W.J. Parkhurst, and R.L. Tanner. 2008. Nighttime chemistry in the Houston urban plume. *Atmospheric Environment* 42:7544-7552.

MacKenzie, J.J., and M.T. El Ashry. 1990. *Air Pollution's Toll on Forests and Crops.* Yale University Press, New Haven, CT. 376 pp.

Mathur, R., and R.L. Dennis. 2003. Seasonal and annual modeling of reduced nitrogen compounds over the eastern United States: Emissions, ambient levels, and deposition amounts. *Journal of Geophysical Research-Atmospheres* 108, 4481, 19 PP., 2003 doi:10.1029/2002JD002794

Mahimairaja, S., N.S. Bolan, M.J. Hedley, and A.N. Macgregor. 1994. Losses and transformation of nitrogen during composting of poultry manure with different amendments - an incubation experiment. *Bioresource Technology* 47:265-273.

Mangiafico, S.S. and K. Guillard. 2006. Fall fertilization timing effects on nitrate leaching and turfgrass color and growth. *Journal of Environmental Quality* 35:163-171.

Malm, W.C., B.A. Schichtel, M.L. Pitchford, L.L. Ashbaugh, and R.A. Eldred. 2004. Spatial and monthly trends in speciated fine particle concentration in the United States. *Journal of Geophysical Research-Atmospheres* 109, D03306, doi:10.1029/2003JD003739.

Maryland Department of the Environment (MDE). 2006. *BNR Costs and Status BNR Project Costs Eligible for State Funding.* Provided by Elaine Dietz on October 31, 2006.

Mathur, R., and R.L. Dennis. 2003. Seasonal and annual modeling of reduced nitrogen compounds over the Eastern United States: Emissions, ambient levels, and deposition amounts. Air Pollution Modeling and its Application XIX, Chapter 4. *Journal of Geophysical Research* 108(D15):ACH 22-1-ACH 22-19.

McCarthy, M.J., P.L. Lavrentyev, L. Yang, L. Zhang, Y. Chen, B. Qin, and W.S. Gardner. 2007. Nitrogen dynamics relative to microbial food web structure in a subtropical, shallow, well-mixed, eutrophic lake (Taihu Lake, China). *Hydrobiologia* 581:195-207.

McCarthy, M.J., R.T. James, Y. Chen, T.L. East, and W.S. Gardner. 2009. Nutrient ratios and phytoplankton community structure in the large, shallow, eutrophic, subtropical Lakes Okeechobee (Florida, USA) and Taihu (China). *Limnology* 10:215-227.

McClenny, W.A., E.J. Williams, R.C. Cohen, and J. Stutz. 2002. Preparing to measure the effects of the NO_x SIP call-methods for ambient air monitoring of NO, NO_2, NO_y, and individual NO_z species. *Journal of the Air and Waste Management Association* 52:542-562.

McMurry, P.H., M.F. Shepherd, and J.S. Vickery. 2004. *Particulate Matter Science for Policy Makers.* Cambridge University Press, Cambridge, UK.

Milesi, C., S. Running, C.D. Elvidge, J.B. Dietz, B.T. Tuttle and R.R. Nemani. 2005. Mapping and modeling the biogeochemical cycling of turf grasses in the United States. *Environmental Management* 36:426-438.

Miller, S.A., A.E. Landis, and T.L. Theis. 2007. Environmental tradeoffs of bio-based production. *Environmental Science and Technology* 41:5176-5182.

Miller, S.A., A.E. Landis, and T.L. Theis. 2006. Use of Monte Carlo analysis to characterize nitrogen fluxes in agroecosystems. *Environmental Science and Technolog,* 40 (7):2324-2332.

Millennium Ecosystem Assessment. 2003. *Ecosystems and Human Well-Being: A Framework For Assessment.* Island Press, Washington, DC.

Miner, J.R., F.J. Humenik, J.M. Rice, D.M.C. Rashash, C.M. Williams, W. Robarge, D.B. Harris, and R. Sheffield. 2003. Evaluation of a permeable, 5 cm thick, polyethylene foam lagoon cover. *Transactions of the American Society of Agricultural and Biological Engineers* 46:1421-1426

Mitsch, W.J., J.W. Day, Jr., J.W. Gilliam, P.M. Groffman, D.L. Hey, G.W. Randall, and N. Wang. 1999. *Reducing Nutrient Loads, Especially Nitrate-Nitrogen, to Surface Water, Groundwater, and the Gulf of Mexico. Topic 5 Report for the Integrated Assessment on Hypoxia in the Gulf of Mexico.* NOAA Coastal Ocean Program Decision Analysis Series No. 19. NOAA Coastal Ocean Program, Silver Spring, MD, 111 p.

Mitsch, W.J., J.W. Day, Jr., J.W. Gilliam, P.M. Groffman, D.L. Hey, G.W. Randall, and N. Wang. 2001. Reducing nitrogen loading to the Gulf of Mexico from the Mississippi River Basin: Strategies to counter a persistent ecological problem. *BioScience* 51:373-388.

Mitsch, W.J. and S.E. Jørgensen. 2004. *Ecological Engineering and Ecosystem Restoration.* John Wiley & Sons, Inc., New York. 411 pp.

Mitsch, W.J., J.W. Day, Jr., L. Zhang, and R. Lane. 2005. Nitrate-nitrogen retention by wetlands in the Mississippi River Basin. *Ecological Engineering* 24:267-278.

Mitsch, W.J., and J.W. Day, Jr. 2006. Restoration of wetlands in the Mississippi-Ohio Missouri (MOM) River Basin: Experience and needed research. *Ecological Engineering* 26: 55-69.

Mitsch, W.J. and J.G. Gosselink. 2007. *Wetlands.* 4th ed., John Wiley & Sons, Inc., New York, 582 p.

Mitsch, W.J., L. Zhang, D.F. Fink, M.E. Hernandez, A.E. Altor, C.L. Tuttle, and A.M. Nahlik. 2008. Ecological engineering of floodplains. *Ecohydrology and Hydrobiology* 8:139-147.

Mitsch, W.J., J.G. Gosselink, C.J. Anderson, and L. Zhang. 2009. *Wetland Ecosystems.* John Wiley & Sons, Inc., New York, 295 p.

Moffit, D.C., and Lander C. 1999. *Using Manure Characteristics to Determine Land-Based Utilization. Natural Resources Conservation Service.* ASAE Paper No. 97-2039, USDA-Natural Resources Conservation Service, Fort Worth, TX, [available at: http://wmc.ar.nrcs.usda.gov/technical/WQ/manurechar.html.]

Mokdad, A.H., J.S. Marks, D.F. Stroup, and J.L. Gerberding. 2004. Actual causes of death in the United States. 2000. *Journal of the American Medical Association* 291:1238-1245.

Moomaw, W.R. and M. Birch. 2005. Cascading costs: An economic nitrogen cycle. *Science in China Series C: Life Science*s 48 Special Issue:678-696.

Morris, J.T. 1991. Effects of nitrogen loading on wetland ecosystems with particular reference to atmospheric deposition. *Annual Reviews of Ecology and Systematics* 22:257-279.

Mosier, A.R. and T. Parkin. 2007. Gaseous Emissions (CO_2, CH_4, N_2O and NO) from diverse agricultural production systems. In *Biodiversity in Agricultural Production Systems,* eds. G. Genckiser and S. Schnell, 317-348. CRC Press, Boca Raton, FL

Mosier, A.R., M.A. Bleken, P. Chaiwanakupt, E.C. Ellis, J.R. Freney, R.B. Howarth, P.A. Matson, K. Minami, R. Naylor, K.N. Weeks and Z.L. Zhu. 2001. Policy implications of human-accelerated nitrogen cycling. Biogeochemistry 52:281-320. Reprinted in *The Nitrogen Cycle at Regional to Global Scales,* eds. E.W. Boyer and R.W. Howarth, 477-516. Kluwer Academic Publishers, Dordrecht.

Moy, L.A., R.R. Dickerson, and W.F. Ryan. 1994. How meteorological conditions affect tropospheric trace gas concentrations in rural Virginia. *Atmospheric Environment* 28(17):2789-2800.

Munger, J.W., S.M. Fan, P.S. Bakwin, M.L. Goulden, A.H. Goldstein, A.S. Colman, and S.C. Wofsy. 1998. Regional budgets for nitrogen oxides from continental sources: Variations of rates for oxidation and deposition with season and distance from source regions, *Journal of Geophysical Research-Atmospheres* 103:8355-8368.

Myhre, G, 2009. Consistency between satellite-derived and modelled estimates of the direct aerosol effect. *Science* 325:187-190.

National Academy of Engineering. 2008. Grand Challenges. http://www.engineeringchallenges.org/cms/challenges.aspx [accessed August 10, 2011]

National Atmospheric Deposition Program. 2010. *National Atmospheric Deposition Program (NADP).* http://nadp.sws.uiuc.edu/

National Research Council. 1976. *Nutrient Requirements of Beef Cattle.* 5th revised ed., National Academies Press, Washington, DC.

National Research Council. 1988. *Nutrient Requirements of Swine.* 9th revised ed., National Academies Press, Washington, DC.

National Research Council.1992. *Restoration of Aquatic Ecosystems.* Committee on Restoration of Aquatic Ecosystems, National Research Council, National Academy Press. Washington, DC

National Research Council.1994. *Nutrient Requirements of Poultry.* 9th revised ed., National Academies Press, Washington, DC.

National Research Council.1996. *Nutrient Requirements of Beef Cattle.* 7th revised ed., National Academies Press, Washington, DC.

National Research Council. 2000. National Research Council. *Clean Coastal Waters: Understanding and Reducing the Effects of Nutrient Pollution.* Ocean Studies Board and Water Science and Technology Board, Commission on Geosciences, Environment, and Resources. National Academy Press, Washington, DC. 405 p.

National Research Council. 2001. *Nutrient Requirements of Dairy Cattle.* 7th revised ed., National Academies Press, Washington, DC.

National Research Council. 2002. *Air Emissions from Animal Feeding Operations: Current Knowledge, Future Trends.* National Academies Press, Washington, DC.

National Research Council. 2003. *Air Emissions from Animal Feeding Operations: Current Knowledge and Future Needs.* National Academies Press, Washington, DC. 263 p.

National Research Council. 2007. *Models in Environmental Regulatory Decision Making,* National Academies Press, Washington DC.

National Research Council. 2008a. *Water Implications of Biofuels Production in the United States.* National Academies Press, Washington, DC.

National Research Council. 2008b. *Mississippi River Water quality and the Clean Water Act: Progress, Challenges, and Opportunities.* National Academies Press, Washington, DC.

National Research Council. 2009. *Nutrient Control Actions for Improving Water Quality in the Mississippi River Basin and the Northern Gulf of Mexico.* National Academies Press, Washington, DC.

Naylor, R, H.W. Steinfeld, J. Falcon, J. Galloway, V. Smil, E. Bradford, J. Alder, and H. Mooney. 2005. Losing the links between livestock and land. *Science* 310:1621-1622.

Neff, J.C., E.A. Holland, F.J. Dentener, W.H. McDowell, and K.M. Russell. 2002. The origin, composition and rates of organic nitrogen deposition: A missing piece of the nitrogen cycle? *Biogeochemistry* 57:99-136.

Nilles, M.A., J.D. Gordon and L.J. Schroder. 1994. The precision of wet atmospheric deposition data from National-Atmospheric-Deposition-Program National-Trends-Network sites determined with collocated samplers. *Atmospheric Environment* 28(6): 1121-1128.

Nilsson, J., and P. Grennfelt. 1988. Critical loads for sulphur and nitrogen. *Environmental Report* 1988:15 (Nord 1988:97), Nordic Council of Ministers, Copenhagen, Denmark. 418 p.

Nixon, S.W. 1995. Coastal marine eutrophication: a definition, social causes, and future concerns. *Ophelia* 41:199-219.

Nolan, B.T., and K.J. Hitt. 2002. *Nutrients in shallow ground waters of beneath relatively undeveloped areas in the conterminous United States:* U.S. Geological Survey Water-Resources Investigations Report 02-4289, 17 p.

Nolan, B.T., and K.J. Hitt. 2006. Vulnerability of shallow groundwater and drinking-water wells to nitrate in the United States. *Environmental Science and Technology* 40(24):7834-7840.

Nolan, B.T., and B.C. Ruddy. 1996. *Nitrate in Ground Waters of the United States – Assessing the Risk.* U.S. Geological Survey Fact Sheet FS-092-96, 4 p.

Nolan, B.T., and J.D. Stoner. 2000. Nutrients in ground waters of the conterminous United States, 1992-1995. *Environmental Science and Technology* 34(7):1156-1165.

North, R.L., S.J. Guildford, R.E.H. Smith, S.M. Havens, and M.R. Twiss. 2007. Evidence for phosphorus, nitrogen, and iron colimitation of phytoplankton communities in Lake Erie. *Limnology and Oceanography* 52:315-328.

Oitjen, J.W., and J.L. Beckett. 1996. Role of ruminant livestock in sustainable agricultural systems. *Journal of Animal Science* 74:1406-1409.

Paerl, H.W. 1982. Factors limiting productivity of freshwater ecosystems. *Advances in Microbial Ecology* 6:75-110.

Paerl, H.W. 1988. Nuisance phytoplankton blooms in coastal, estuarine, and inland waters. *Limnology and Oceanography* 33:823 847.

Paerl, H.W. 1997. Coastal eutrophication and harmful algal blooms: Importance of atmospheric deposition and groundwater as "new" nitrogen and other nutrient sources. *Limnology and Oceanography* 42:1154-1165.

Paerl, H.W. 2009. Controlling Eutrophication along the freshwater–marine continuum: Dual nutrient (N and P) reductions are essential. *Estuaries and Coasts* 32:593-601.

Paerl, H.W., and D.R. Whitall. 1999. Anthropogenically-derived atmospheric nitrogen deposition, marine eutrophication and harmful algal bloom expansion: Is there a link? *AMBIO: A Journal of the Human Environment* 28:307-311.

Paerl, H.W., R.L. Dennis, and D.R. Whitall. 2002. Atmospheric deposition of nitrogen: Implications for nutrient over-enrichment of coastal waters. *Estuaries* 25:677-693.

Paerl, H.W., L.M. Valdes, A.R. Joyner, and V. Winkelmann. 2007. Phytoplankton indicators of ecological change in the eutrophying Pamlico Sound system, North Carolina. *Ecological Applications* 17: S88-S101.

Park, R.J., K.E. Pickering, D.J. Allen, G.L. Stenchikov, and M.S. Fox-Rabinovitz. 2004a. Global simulation of tropospheric ozone using the University of Maryland Chemical Transport Model (UMD-CTM): 2. Regional transport and chemistry over the central United States using a stretched grid. *Journal of Geophysical Research-Atmospheres.* 109, D09303, doi:10.1029/2003JD004269.

Parkin, T.B., and T.C. Kaspar. 2006. Nitrous Oxide Emissions from Corn-Soybean Systems in the Midwest. *Journal of Environmental Quality* 35:1496-1506.

Parrish, D.D., and F.C. Fehsenfeld. 2000. Methods for gas-phase measurements of ozone, ozone precursors and aerosol precursors. *Atmospheric Environment* 34:1921-1957.

Parrish, D.D., M.P. Buhr, M. Trainer, R.B. Norton, J.P. Shimshock, F.C. Fehsenfeld, K.G.Anlauf, J.W. Bottenheim, Y.Z. Tang, H.A. Wiebe, J.M. Roberts, R.L. Tanner, L. Newman, V.C. Bowersox, K.J. Olszyna, E.M. Bailey, M.O. Rodgers, T. Wang, H. Berresheim, U.K. Roychowdhury, and K.L. Demerjian 1993. The total reactive oxidized nitrogen levels and the partitioning between the individual-species at 6 rural sites in Eastern North-America. *Journal of Geophysical Research-Atmospheres* 98:2927-2939.

Parrish, D.D., Y. Kondo, O.R. Cooper, C.A. Brock D.A. Jaffe, M. Trainer, T. Ogawa, G. Hübler, and F.C. Fehsenfeld. 2004a. Intercontinental transport and chemical transformation 2002 (ITCT 2K2) and Pacific Exploration of Asian Continental Emission (PEACE) experiments: An overview of the 2002 winter and spring intensives. *Journal of Geophysical Research-Atmospheres* 109, D23S01, doi:10.1029/2004JD004980.

Parrish, D.D., T.B. Ryerson, J.S. Holloway, J.A. Neuman, J.M. Roberts, J. Williams, C.A. Stroud, G.J. Frost, M. Trainer, G. Hubler, F.C. Fehsenfeld, F. Flocke, and A.J. Weinheimer. 2004b. Fraction and composition of NO_y transported in air masses lofted from the North American continental boundary layer. *Journal of Geophysical Research-Atmospheres* 109, D09302, doi:10.1029/2003JD004226.

Peierls, B.L., N.F. Caraco, M.L. Pace, and J.J. Cole. 1991. Human influence on river nitrogen. *Nature* 350:386-387.

Penner, J.E., C.S. Atherton, J. Dignon, S.J. Ghan, J.J. Walton, and S. Hameed. 1991. Tropospheric nitrogen - a 3-dimensional study of sources, distributions, and deposition. *Journal of Geophysical Research-Atmospheres* 96:959-990.

Peoples, M.B., J.R. Freney and A.R. Mosier. 1995. Minimizing gaseous losses of nitrogen. In *Nitrogen Fertilization in the Environment,* ed. P.E. Bacon, 565-602. Marcel Dekker, Inc. New York, NY.

Peoples M.B., E.W. Boyer, K.W.T. Goulding, P. Heffer, V.A. Ochwoh, B. Vanlauwe, S. Wood, K. Yagi, and O. Van Cleemput. 2004. Pathways of nitrogen loss and their impacts on human health and the environment. In *Agriculture and the nitrogen cycle: assessing the impact of fertilizer use on food production and the environment,* eds. A.R. Mosier, K. Syers and J.R. Freney, 53-69. Washington, D.C. Island Press.

Petrovic, A.M. 1990. The fate of nitrogenous fertilizers applied to turfgrass. *Journal of Environmental Quality* 19:1-14.

Petrovic, A.M. 2004a. Nitrogen source and timing impact on nitrate leaching from turf. In *1st IC on Turfgrass,* ed. P.A. Nektarios, 427-432. Acta Hort. 661. ISHS.

Petrovic, A.M. 2004b. Managing sports fields to reduce environmental impacts. In *1st IC on Turfgrass,* ed. P.A. Nektarios, 405-412. Acta Hort. 661. ISHS.

Petrovic, A.M., and I.M. Larsson-Kovach. 1996. Effect of maturing turfgrass soils on the leaching of the herbicide mecoprop. *Chemosphere* 33:585-593.

Piehler, M. F., J. Dyble, P.H. Moisander, J.L.K. Pinckney, and H.W. Paerl. 2002. Effects of modified nutrient concentrations and ratios on the structure and function of the native phytoplankton community in the Neuse River Estuary, North Carolina USA. *Aquatic Ecology* 36:371-385.

Pinckney, J.L., H.W. Paerl, and M.B. Harrington. 1999. Responses of the phytoplankton community growth rate to nutrient pulses in variable estuarine environments. *Journal of Phycology* 35:1455-1463.

Pinder, R.W., A.B. Gilliland, and R.L. Dennis. 2008. Environmental impact of atmospheric NH_3 emissions under present and future conditions in the eastern United States. *Geophysical Research Letters* 35, L12808, doi:10.1029/2008GL033732.

Pope C.A. 2000a. Epidemiology of fine particulate air pollution and human health: biologic mechanisms and who's at risk? *Environmental Health Perspectives* 108 Suppl 4:713-723.

Pope C.A. 2000b. What do epidemiologic findings tell us about health effects of environmental aerosols? *Journal of Aerosol Medicine* 13(4):335-354.

Pope, C.A., M. Thun, M. Namboodiri, D. Dockery, J. Evans, F. Speizer, and C. Heath. 1995. Particulate air pollution as a predictor of mortality in a prospective study of U.S. adults. *American Journal Respiratory Critical Care Medicine* 151:669-674.

Pope, C.A., M. Ezzati, and D.W. Dockery. 2009. Fine-Particulate Air Pollution and Life Expectancy in the United States. *New England Journal of Medicine* 360:376-86.

Poulida, O., K.L. Civerolo, and R.R. Dickerson. 1994. Observations and Tropospheric Photochemistry in Central North-Carolina. *Journal of Geophysical Research-Atmospheres* 99:10553-10563.

Powers, S. 2007. Nutrient loads to surface water from row crop production. *The International Journal of Life Cycle Assessment* 12 (6):399-407.

Prospero, J.M., M. Uematsu, and D.L. Savoie. 1989. Mineral aerosol transport to the Pacific Ocean. In *Chemical Oceanography,* eds. R. Chester and R.A. Duce, 187-218. Academic Press, London

Prospero, J.M., K. Barrett, T. Church, F. Dentener, R.A. Duce, J.N. Galloway, H. Levy II, J. Moody, and P. Quinn.1996. Atmospheric deposition of nutrients to the North Atlantic Basin. *Biogeochemistry* 35:27-73.

Rabalais, N.N., and R.E. Turner (eds). 2001. *Coastal Hypoxia: Consequences for Living Resources and Ecosystems.* Coastal and Estuarine Studies 58. American Geophysical Union, Washington, DC. 454p.

Rabalais, N.N., R.E. Turner, D. Justic, Q. Dortch, W.J. Wiseman Jr., and B.K. Gupta. 1996. Nutrient changes in the Mississippi River and system responses on the adjacent continental shelf. *Estuaries* 19:386-407.

Rabalais, N. N., R.E. Turner, D. Justic, Q. Dortch, and W. J. Wiseman, Jr. 1999. *Characterization of Hypoxia. Topic 1 Report for the Integrated Assessment of Hypoxia in the Gulf of Mexico.* National Oceanic and Atmospheric Administration Coastal Ocean Program Decision Analysis Series No. 15. NOAA Coastal Ocean Program, Silver Spring, Maryland.

Ribaudo, M.O., and C.J. Nickerson. 2009. Agriculture and water quality trading: Exploring the possibilities. *Journal of Soil and Water Conservation* 64(1):1-7.

Ridley, B.A., J.G. Walega, J.E. Dye, and F.E. Grahek.1994. Distribution of NO, NO_x, NO_y, and O_3 to 12 km altitude during the summer monsoon season over New Mexico. *Journal of Geophysical Research* 99: 25519-25534.

Riegman, R. 1998. Species composition of harmful algal blooms in relation to macronutrient dynamics. In *Physiological ecology of Harmful Algal Blooms,* eds. D.M. Anderson, A.D. Cembella, and G.M. Hallegraeff, 475-488. NATO Series Vol. G 41.

Robertson G., V.H. Dale, O.C. Doering, S.P. Hamburg, J.M. Melillo, M.M. Wander, W. J. Parton, P.R. Adler, J.N. Barney, R.M. Cruse, C.S. Duke, P.M. Fearnside, R.F. Follett, H.K. Gibbs, J. Goldemberg, D. J. Mladenoff, D. Ojima, M.W. Palmer, A. Sharpley, L. Wallace, K.C. Weathers, J.A. Wiens, and W.W. Wilhelm. 2008. Sustainable biofuels redux. *Science* 322(5898):49-50.

Rupert, M.G. 2008. Decadal-scale changes of nitrate in ground water of the United States, 1988-2004. *Journal of Environmental Quality* 37:S-240-S-248.

Ryan, W. F., B.G. Doddridge, R.R. Dickerson, R.M. Morales, K.A. Hallock, P.T. Roberts, D.L. Blumenthal, J.A. Anderson, and K.L. Civerolo. 1998. Pollutant transport during a regional O_3 episode in the Mid-Atlantic States. *Journal of the Air and Waste Management Association* 48(9):786-797.

Ryther, J., and W. Dunstan. 1971. Nitrogen, phosphorus and eutrophication in the coastal marine environment. *Science* 171:1008-1112.

Salvagioti F., K.G. Cassman, J.E. Specht, D.T. Walters, A. Weiss, and A. Dobermann. 2008. Nitrogen uptake, fixation and response to N fertilizer in soybeans: A review. *Field Crops Research* 108:1-13.

Sawyer, J., E. Nafziger, G. Randall, L. Bundy, G. Rehm, and B. Joern. 2006. *Concepts and Rationale for Regional Nitrogen Rate Guidelines for Corn. Iowa State Extension PM 2015.* www.extension.iastate.edu/Publications/2015.pdf [accessed April 29, 2010]

Schindler, D.W. 1971. Carbon, nitrogen, and phosphorus and the eutrophication of freshwater lakes. *Journal of Phycology* 7:321-329.

Schindler, D.W., R.E. Hecky, D.L. Findlay, M.P. Stainton, B.R. Parker, M. Paterson, K.G. Beaty, M. Lyng, and S.E.M. Kasian. 2008. Eutrophication of lakes cannot be controlled by reducing nitrogen input: Results of a 37 year whole ecosystem experiment. *Proceedings of the National Academy of Sciences* 105:11254-11258.

Schlesinger, W.H. 2009. On the fate of anthropogenic nitrogen. *Proceedings of the National Academy of Sciences* 106:203-208.

Schwab, J.J., J.B. Spicer, and K.L. Demerjian. 2009. Ozone, trace gas, and particulate matter measurements at a rural site in southwestern New York State: 1995-2005. *Journal of the Air and Waste Management Association* 59:293-309.

Schwede, D., R. Dennis, and M. Bitz. 2009. The Watershed Deposition Tool: A tool for incorporating atmospheric deposition in water-quality analyses. *Journal of the American Water Resources Association* 45(4):973-985. (Online URL for tool: htttp://www.epa.gov/amad/EcoExposure/depositionMapping.html) [accessed July 9, 2009]

Scott, B., R.M. Peck, C. Tallarico, and J.A. Kostel, J.A. (In preparation). *Nitrogen Farming in the Illinois River Watershed: An Environmental Economic Market Comparison.*

Scott, J.T., R.D. Doyle, S.J. Prochnow, and J.D. White. 2008. Are watershed and lacustrine controls on planktonic nitrogen fixation hierarchically structured? *Ecological Applications* 18:805-819.

Scott, J.T., J.K. Stanley, R.D. Doyle, M.G. Forbes, and B.W. Brooks. 2009. River-reservoir transition zones are nitrogen fixation hot spots regardless of ecosystem trophic state. *Hydrobiologia* 625: 61-68.

Seinfeld, J. H. and S.N, Pandis. 1998. *Atmospheric Chemistry and Physics,* Wiley, New York, NY.

Seitzinger, S.P., and Giblin, A.E. 1996. Estimating denitrification in North Atlantic continental shelf sediments. *Biogeochemistry* 35:235-259.

Selman, M., S. Greenhalgh, R. Diaz and Z. Sugg. 2008. *Eutrophication and Hypoxia in Coastal Areas: a Global Assessment of the State of Knowledge.* WRI Policy Note, Water Quality: Eutrophication and Hypoxia. No.1. World Resources Inst., Washington, DC. 6 p.

Shores, R.C., D.B. Harris, E.L. Thompson, Jr., C.A. Vogel, D. Natschke, R.A. Hashmonay, K.R. Wagoner, and M. Modrak. 2005. Plane-integrated open-path fourier transform infrared spectrometry methodology for anaerobic swine lagoon emission measurements. A*pplied Engineering in Agriculture* 21:487-492.

Shuman, L.M. 2002. Phosphorus and nitrate nitrogen in runoff following fertilizer application to turfgrass. *Journal of Environmental Quality* 31:1710-1715.

Sickles, J., and D.S. Shadwick. 2007a. Changes in air quality and atmospheric deposition in the eastern United States: 1990-2004. *Journal of Geophysical Research-Atmospheres* 112(D17), CiteID D17301112.

Sickles, J.E., and D.S. Shadwick. 2007b. Seasonal and regional air quality and atmospheric deposition in the eastern United States. *Journal of Geophysical Research-Atmospheres* 112, D17302, doi:10.1029/2006JD008356.

Slater, J.V., D.L. Terry, and B.J. Kirby. 2010. *Commercial Fertilizers* 2008. Association of American Plant Food Control Officials, Inc., Lexington, KY

Smil, V. 1999. Nitrogen in crop production: An account of global flows. *Global Biogeochemical. Cycles* 13:647-662.

Smil, V. 2001. *Enriching the Earth: Fritz Haber, Carl Bosch and the Transformation of World Food Production.* MIT Press, Cambridge, MA

Smith, R.A., G.E. Schwarz, and R.B. Alexander. 1997. Regional interpretation of water-quality monitoring data. *Water Resources Research* 33:2781-2798.

Smith, V.H. 1990. Nitrogen, phosphorus, and nitrogen fixation in lacustrine and estuarine ecosystems. *Limnology and Oceanography* 35:1852-1859.

Snyder, C.S., T.W. Bruulsema, and T.L. Jensen. 2007. *Greenhouse Gas Emissions from Cropping Systems and the Influence of Fertilizer Management - a Literature Review.* International Plant Nutrition Institute, Norcross, GA.

Sokolov, A.P., D.W. Kicklighter, J.M. Melillo, B.S. Felzer, C.A. Schlosser, and T.W. Cronin 2008. Consequences of considering carbon-nitrogen interactions on the feedbacks between climate and the terrestrial carbon cycle. *Journal of Climate* 21:3776-3796.

Solley, W.B., R.R. Pierce, and H.H. Perlman. 1998. *Estimated use of water in the United States in 1995.* U.S. Geological Survey Circular 1200. U.S. Geological Survey.

Sommariva, R., M.Trainer, J.A. de Gouw, J.M. Roberts, C. Warneke, E. Atlas, F. Flocke, P.D. Goldan, W.C. Kuster, A.L. Swanson, and F.C. Fehsenfeld. 2008. A study of organic nitrates formation in an urban plume using a Master Chemical Mechanism. *Atmospheric Environment* 42(23):5771-5786.

Sommer, S.G. 1997. Ammonia volatilization from farm tanks containing anaerobically digested animal slurry. *Atmospheric Environment* 31:863-868.

State-EPA Nutrient Innovations Task Group. 2009. *An Urgent Call to Action: Report of the State-EPA Nutrient Innovations Task Group.* www.epa.gov/waterscience/criteria/nutrient/nitgreport.pdf [accessed August, 19, 2010]

Stephen, K., and V.P. Aneja. 2008. Trends in agricultural ammonia emissions and ammonium concentrations in precipitation over the Southeast and Midwest United States. *Atmospheric Environment* 42, 3238-3252.

Stolte, W., T. McCollin, A. Noodeloos, and R. Riegman, 1994. Effects of nitrogen source on the size distribution within marine phytoplankton populations. *Journal of Experimental Marine Biology and Ecology* 184: 83-97.

Sullivan, L.J., T.C. Moore, V.P. Aneja, W.P. Robarge, T.E. Pierce, C. Geron and B. Gay, 1996. Environmental variables controlling nitric oxide emissions from agricultural soils in the southeast United States. *Atmospheric Environment* 30:3573-3582.

Sutton, M.A., W.A.H. Asman, T. Ellermann, J.A. Van Jaarsveld, K. Acker, V.P. Aneja, J. Duyzer, L. Horvath, S. Paramonov, M. Mitosinkova, Y.S. Tang, B. Ackermann, T. Gauger, J. Bartniki, A. Neftel, and J.W. Erisman. 2003. Establishing the link between ammonia emission control and measurements of reduced nitrogen concentrations and deposition. *Journal of Environmental Monitoring and Assessment* 82:149-185.

Sutton, M.A., E. Nemitz, J.W. Erisman, C. Beier, K B. Bahl, P. Cellier, W. de Vries, F. Cotrufo, U. Skiba, C. Di Marco, S. Jones, P. Laville, J.F. Soussana, B. Loubet, M. Twigg, D. Famulari, J. Whitehead, M.W. Gallagher, A. Neftel, C.R. Flechard, B. Herrmann, P.L. Calanca, J.K. Schjoerring, U. Daemmgen, L. Horvath, Y.S. Tang, B.A. Emmett, A. Tietema, J. Penuelas, M. Kesik, N. Brueggemann, K. Pilegaard, T. Vesala, C.L. Campbell, J.E. Olesen, U. Dragosits, M.R. Theobald, P. Levy, D.C. Mobbs, R. Milne, N. Viovy, N. Vuichard, J.U. Smith, P. Smith, P. Bergamaschi, D. Fowler, and S. Reis. 2007. Challenges in quantifying biosphere-atmosphere exchange of nitrogen species. *Environmental Pollution* 150:125-139.

Syrett, P.J. 1981. Nitrogen metabolism of microalgae. *Canadian Bulletin of Fisheries and Aquatic Sciences* 210:182-210.

Takegawa, N., Y. Kondo, M. Koike, M. Ko, K. Kita, D.R. Blake, N. Nishi, W. Hu, J B. Liley, S. Kawakami, T. Shirai, Y. Miyazaki, H. Ikeda, J. Russel-Smith, and T. Ogawa. 2003. Removal of NO_x and NO_y in biomass burning plumes in the boundary layer over northern Australia. *Journal of Geophysical Research-Atmospheres* 108, D10 4308, doi: 10.1029/2002JD002505

Templeton, S.R., D. Zilberman, and S.J. Yoo. 1998. An economic perspective on outdoor residential pesticide use. *Environmental Science & Technology* 32: 416A–423A.

Terry, D.L., and B.J. Kirby. 2006. *Commercial Fertilizers.* Association of American Plant Food Control Officials (AAPFCO), Lexington, KY.

Thornton, P.E., J.F. Lamarque, N.A. Rosenbloom, and N.M. Mahowald. 2007. Influence of carbon-nitrogen cycle coupling on land model response to CO_2 fertilization and climate variability. *Global Biogeochemical Cycles* 21:GB4018, doi:10.1029/2006GB002868.

TWI. 2007. *Assessing Tax Impacts of Nutrient Management Options: Nutrient Farming Could Lessen Tax Burden to Chicago Area Residents.* The Wetlands Initiative, Chicago, IL.

U.S. Department of Agriculture. 2002. *Census of Agriculture.* www.agcensus.usda.gov/

U.S. Department of Agriculture. 2007. *Census of Agriculture.* www.agcensus.usda.gov/

U.S. Department of Agriculture. 2008. *Fertilizer Use and Price.* www.ers.usda.gov/data/fertilizeruse

U.S. Department of Agriculture Economic Research Service. 2006. *Agricultural Resources Environmental Indicators,* eds. K. Weibe and N. Gollehon, Economic Information Bulletin No. 16, Economic Research Service, Washington, DC.

U.S. Department of Agriculture National Agricultural Statistics Service. 2007. *Data and Statistics.* www.nass.usda.gov/Data_and_Statistics/

U.S. Department of Agriculture National Agricultural Statistics Service. 2008. Crop Production Historical Track Records, April 2008. U.S. Department of Agriculture National Agricultural Statistics Service. 224 p. [available at: usda.mannlib.cornell.edu/usda/nass/htrcp//2000s/2008 htrcp-04-30-2008.pdf]

U.S. Department of Agriculture Natural Resources Conservation Service. 2007. *A Statistical Survey of Land Use and Natural Resource Conditions and Trends on U.S. Non-Federal Lands.* www.nrcs.usda.gov/technical/NRI/

U.S. Environmental Protection Agency. 1986. *Quality Criteria for Water 1986.* EPA 440/5-86-001. U.S. Environmental Protection Agency, Office of Water, Washington, DC. 477 p.

U.S. Environmental Protection Agency. 1997. *National Water Quality Inventory 1996 Report to Congress.* EPA841-R-97-006, U.S. Environmental Protection Agency, Washington, DC.

U.S. Environmental Protection Agency. 1998. *Regulatory Impact Analysis for the NO_x SIP Call, FIP, and Section 126 Petitions.* U.S. Environmental Protection Agency, Washington, DC.

U.S. Environmental Protection Agency. 1999. *1999 Update of Ambient Water Quality Criteria for Ammonia.* EPA 822-R-99-014, U.S. Environmental Protection Agency, Office of Water, Washington, DC. 153 p.

U.S. Environmental Protection Agency. 2000a. *National Water Quality Inventory: 1998 report to Congress.* EPA-841-R-00-001, U.S. EPA, Office of Water, Washington, DC. 434 p.

U.S. Environmental Protection Agency. 2000b. *Nutrient Criteria Technical Guidance Manual: Rivers and Streams.* EPA-822-B-00-002, Environmental Protection Agency, Office of Water, Office of Science and Technology, Washington, DC 20460.

U.S. Environmental Protection Agency. 2000c. *Nutrient Criteria Technical Guidance Manual. Lakes and Reservoirs.* EPA-822-B-00-001, U.S. EPA, Office of Water, Office of Science and Technology, Washington, DC. 232 p.

U.S. Environmental Protection Agency. 2001a. *National Coastal Condition Report.* EPA-620/R-01/005, U.S. Environmental Protection Agency, Office of Research and Development and Office of Water, Washington, DC.

U.S. Environmental Protection Agency. 2001b. *Nutrient Criteria Technical Guidance Manual. Estuaries and Coastal Marine Waters.* EPA-822-B-01-003, U.S. Environmental Protection Agency, Office of Water, Washington, DC. [available at: epa.gov/waterscience/criteria/nutrient/guidance/index.html]

U.S. Environmental Protection Agency. 2002. *National Water Quality Inventory: 2000 Report.* EPA-841-R-02-001, U.S. Environmental Protection Agency, Office of Water, Washington, DC.

U.S. Environmental Protection Agency. 2004. *National Coastal Condition Report II.* EPA 620/R-03/002, U.S. Environmental Protection Agency, Office of Research and Development and Office of Water, Washington, DC.

U.S. Environmental Protection Agency. 2005a. *Air Quality Criteria for Particulate Matter,* EPA/600/P-99/002aF, U.S. Environmental Protection Agency, Washington, DC.

U.S. Environmental Protection Agency. 2005b. *National Emissions Inventory.* United States Environmental Protection Agency, Washington D.C. [available at: www.epa.gov/ttn/chief/net/2002inventory.html]

U.S. Environmental Protection Agency. 2005c. *Regulatory Impact Analysis for the Final Clean Air Interstate Rule.* EPA-452/R-05-002, U.S. Environmental Protection Agency, Washington, DC.

U.S. Environmental Protection Agency. 2006a. *Air Quality Criteria for Ozone and Related Photochemical Oxidants.* EPA/600/R-05/0004aA, U.S. Environmental Protection Agency, Washington, DC.

U.S. Environmental Protection Agency. 2006b. *National Estuary Program Coastal Condition Report.* EPA-842/B-06/001, U.S. Environmental Protection Agency, Office of Water, Office of Research and Development, Washington, DC. 445 p. [available at: www.epa.gov/owow/oceans/nccr/]

U.S. Environmental Protection Agency. 2006c. *Wadeable Streams Assessment. A Collaborative Survey of the Nation's Streams.* EPA-841-B-06-002, U.S. Environmental Protection Agency, Office of Water, Office of Research and Development, Washington, DC. 98 p. [available at: www.epa.gov/owow/streamsurvey/]

U.S. Environmental Protection Agency. 2006d. *2006-2011 EPA Strategic Plan: Charting our Course.* EPA-190-R-06-001, U.S. Environmental Protection Agency, Washington, DC

U.S. Environmental Protection Agency. 2007a. *Annexes for the Integrated Science Assessment for Oxides of Nitrogen – Health Criteria.* Draft Review Report EPA/600/R-07/093, U.S. Environmental Protection Agency, Office of Research and Development, National Center for Environmental Assessment, Washington, DC.

U.S. Environmental Protection Agency. 2007b. *Memorandum: Nutrient Pollution and Numeric Water Quality Standards. From: Benjamin H. Grumbles, Assistant Administrator, U.S. Environmental Protection Agency Office of Water.* May 25, 2007. [available at: www.epa.gov/waterscience/criteria/nutrient/files/policy20070525.pdf]

U.S. Environmental Protection Agency. 2007c. *Nutrient Criteria Technical Guidance Manual. Wetlands.* EPA-822-R-07-004, U.S. Environmental Protection Agency, Office of Water, Office of Science and Technology, Washington, DC. 197 p. [available at: http://epa.gov/waterscience/criteria/nutrient/guidance/index.html]

U.S. Environmental Protection Agency, 2007d. *Summary Report of Air Quality Modeling Research Activities for 2006.* EPA /600/R-07/103, U.S. Environmental Protection Agency, Washington, DC.

U.S. Environmental Protection Agency. 2007e. *U.S. Environmental Protection Agency. Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990-2005.* EPA 430-R-07-002, U.S. Environmental Protection Agency, Washington DC.

U.S. Environmental Protection Agency. 2007f. Biological Nutrient Removal Processes and Costs. EPA-823-R-07-002, U.S. Environmental Protection Agency, Office of Water, Washington, DC. 15 p.

U.S. Environmental Protection Agency. 2008a. *Acid Rain.* www.epa.gov/acidrain/. [accessed May 19, 2009]

U.S. Environmental Protection Agency. 2008b. *CWNS 2008 Report to Congress.* http://water.epa.gov/scitech/datait/databases/cwns/2008reportdata.cfm

U.S. Environmental Protection Agency. 2008c. *EPA's 2006 Report on the Environment.* EPA-600-R-07-045F, U.S. Environmental Protection Agency, Washington, DC.

U.S. Environmental Protection Agency. 2008d. *Integrated Science Assessment for Nitrogen and Sulfur Oxides: Ecological Criteria. (Welfare-based) National Ambient Air Quality Standards (NAAQS).* EPA/600/R-08/082F, U.S. Environmental Protection Agency, Washington, DC.

U.S. Environmental Protection Agency. 2008e. *Municipal Nutrient Removal Technologies Reference Document. Vol. 1 – Technical Report.* EPA 832-R-08-006, U.S. Environmental Protection Agency Office of Wastewater Management, Washington, DC. 268 p.

U.S. Environmental Protection Agency. 2008f. *Municipal Nutrient Removal Technologies Reference Document. Vol. 2 – Appendices.* EPA 832-R-08-00, U.S. Environmental Protection Agency, Office of Water, Office of Wastewater Management, Washington, DC. 181 p.

U.S. Environmental Protection Agency. 2009a. *EPA's Ecosystem Services Research Program 2009-2014: Conserving Ecosystem Services through Proactive Decision Making.* Presentation to the Science Advisory Board Meeting on Strategic Research Directions, April 23-24, 2009.

U.S. Environmental Protection Agency. 2009b. *Draft 2009 Update. Aquatic Life Ambient Water Quality Criteria for Ammonia – Freshwater.* EPA 822-D-09-001, U.S. Environmental Protection Agency, Office of Water, Washington, DC. 192 p.

U.S. Environmental Protection Agency. 2009c. *Continuous Emission Monitoring - Information, Guidance, etc.* www.epa.gov/ttnemc01/cem.html

U.S. Environmental Protection Agency. 2010a. *National Trends in Nitrogen Dioxide Levels.* www.epa.gov/air/airtrends/nitrogen.html

U.S. Environmental Protection Agency. 2010b. *Clean Air Markets. Annual ARP Coal-fired Power Plant Emission Data: 2008 vs. 2009.* www.epa.gov/airmarkets/quarterlytracking.html [accessed April 27, 2010]

U.S. Environmental Protection Agency. 2010c. *National Assessment Database.* www.epa.gov/waters/305b/index.html [accessed April 27, 2010]

U.S. Environmental Protection Agency. 2010d. *Water Quality Reports* http://water.epa.gov/type/watersheds/monitoring/reporting.cfm [accessed April 27, 2010]

U.S. Environmental Protection Agency. 2010e. *Watershed Assessment, Tracking & Environmental Results.* http://iaspub.epa.gov/waters10/attains_nation_cy.control?p_report_type=T#causes_303d [accessed April 27, 2010]

U.S. Environmental Protection Agency and U.S. Department of Agriculture. 1998. *Clean Water Action Plan: Restoring and Protecting America's Waters.* EPA840-R-98-001, U.S. Environmental Protection Agency and U.S Department of Agriculture, Washington, DC. 89 p.

U.S. Environmental Protection Agency Clean Air Scientific Advisory Committee. 2008. *Clean Air Scientific Advisory Committee Recommendations Concerning the Final Rule for the National Ambient Air Quality Standards for Ozone.* EPA-CASAC-08-009, U.S. Environmental Protection Agency, Washington, DC.

U.S. Environmental Protection Agency Clean Air Scientific Advisory Committee. 2009. *Consultation on Particulate Matter National Ambient Air Quality Standards: Scope and Methods Plan for Urban Visibility Impact Assessment.* EPA-CASAC-09-010, U.S. Environmental Protection Agency, Washington, DC.

U.S. Environmental Protection Agency Science Advisory Board. 1973. *Nitrogenous Compounds in the Environment.* EPA-SAB-73-001, U.S. Environmental Protection Agency Science Advisory Board, Washington, DC.

U.S. Environmental Protection Agency Science Advisory Board. 2000. *Toward Integrated Environmental Decision Making.* EPA-SAB-EC-00-001, U.S. EPA Science Advisory Board, Washington, DC

U.S. Environmental Protection Agency Science Advisory Board. 2007. *Hypoxia in the Northern Gulf of Mexico: An Update by the EPA Science Advisory Board.* EPA-SAB-08-003, U.S. Environmental Protection Agency Science Advisory Board, Washington, DC.

U.S. Environmental Protection Agency Science Advisory Board. 2008. *SAB Advisory on the EPA Ecological Research Program Multi-Year Plan.* EPA-SAB-08-011, U.S. Environmental Protection Agency, Office of the Science Advisory Board, Washington, DC.

U.S. Environmental Protection Agency Science Advisory Board. 2009. *Valuing the Protection of Ecological Systems and Services: A Report of the EPA Science Advisory Board.* EPA-SAB-09-012, U.S. Environmental Protection Agency Science Advisory Board, Washington, DC.

Vallino, J.J., C.S. Hopkinson, and J.E. Hobbie. 1996. Modeling bacterial utilization of dissolved organic matter: optimization replaces Monod growth kinetics. *Limnology and Oceanography* 41:1591-1609

Veldkamp, E. and M. Keller. 1997. Fertilizer-induced nitric oxide emissions from agricultural soils. *Nutrient Cycling in Agroecosystems* 48:69-77.

Valdes-Weaver LM, M.F. Piehler, J.L. Pinckney, K.E. Howe. K. Rosignol, and H.W. Paerl. 2006. Long-term temporal and spatial trends in phytoplankton biomass and class-level taxonomic composition in the hydrologically variable Neuse-Pamlico estuarine continuum, NC, USA. *Limnology and Oceanography* 51:1410-1420

Valigura, R.A., R.B. Alexander, M.S. Castro, T.P. Meyers, H.W. Paerl, P.E. Stacey and R.E. Turner (eds.). 2001. *Nitrogen loading in coastal water bodies. An atmospheric perspective. Coastal and Estuarine Studies,* American Geophysical Union, Washington, DC. 254 p.

Van Breemen, N, E.W. Boyer, C.L. Goodale, N.A. Jaworski, K. Paustian, S.P. Seitzinger, K. Lajtha, B. Mayer, D. VanDam, R.W. Howarth, K.J. Nadelhoffer, M. Eve, and G. Billen 2002. Where did all the nitrogen go? Fate of nitrogen inputs to large watersheds in the northeastern USA. *Biogeochemistry* 57:267-293.

Vanni, M.J., W.H. Renwick, J.L. Headworth, J.D. Auch, and M.H. Schaus. 2001. Dissolved and particulate nutrient flux from three adjacent agricultural watersheds: A five-year study. *Biogeochemistry* 54(1):85-114.

Verhoeven, J.T.A., B. Arheimer, C. Yin, and M.M. Hefting. 2006. Regional and global concerns over wetlands and water quality. *Trends in Ecology and Evolution* 21: 96-103.

Verity, P.G., M. Alber, and S.B. Bricker. 2006. Development of hypoxia in well-mixed subtropical estuaries in the Southeastern USA. *Estuaries and Coasts* 29:665-673

Verma, S.B., A. Dobermann, K.G. Cassman, D.T. Walters, J.M. Knops, T.J. Arkebauer, A.E. Suyker, G.G. Burba, B. Amos, H.S. Yang, D. Ginting, K.G. Hubbard, A.A.Gitelson, and E.A.Walter-Shea. 2005. Annual carbon dioxide exchange in irrigated and rainfed maize-based agroecosystems. *Agricultural and Forest Meteorology* 131:77-96.

Vinlove, F.K., and R.F. Torla. 1995. Comparative estimations of U.S. home lawn area. *Journal of Turfgrass Management* 1:83-97.

Vitousek, P.M., J. Aber, R.W. Howarth, G.E. Likens, P.A. Matson, D.W. Schindler, W.H. Schlesinger, and D.G. Tilman. 1997a. Human alteration of the global nitrogen cycle: Causes and Consequences. *Issues in Ecology* 1:1-15.

Vitousek, P.M., H.A. Mooney, J. Lubchenko, J., and J.M. Mellilo. 1997b. Human domination of Earth's ecosystem. *Science* 277: 494-499.

Vollenweider, R.A. 1992. Coastal marine eutrophication: principles and control. *Science of the Total Environment 126(Supplement):1-20.*

Vollenweider, R.A., R. Marchetti, and R.Viviani (eds).1992. *Marine Coastal Eutrophication.* Elsevier Science, New York, NY.

Wang, H.Q., D. J. Jacob, P. LeSager, D.G. Streets, R.J. Park, A.B. Gilliland, and A. vanDonkelaar. 2009. Surface ozone background in the United States: Canadian and Mexican pollution influences. *Atmospheric Environment* 43(6):1310-1319

Wang, N., C.G. Ingersoll , D.K. Hardesty, I.E. Greer, D.J. Hardesty, C.D. Ivey, J.L. Kunz, W.G. Brumbaugh, F.J. Dwyer, A.D. Roberts, J.T. Augspurger, C.M. Kane, R.J. Neves and M.C. Barnhart. 2007a. Contaminant sensitivity of freshwater mussels: Chronic toxicity of copper and ammonia to juvenile freshwater mussels (Unionidae). *Environmental Toxicology and Chemistry* 26(10):2048-2056

Wang, N., C.G. Ingersoll , D.K. Hardesty, C.D. Ivey, J.L. Kunz, T.W. May, F.J. Dwyer, A.D. Roberts, T. Augspurger, C.M. Kane, R.J. Neves and M.C. Barnhart. 2007b. Contaminant sensitivity of freshwater mussels: Acute toxicity of copper, ammonia, and chlorine to glochidia and juveniles of freshwater mussels (Unionidae). *Environmental Toxicology and Chemistry* 26(10):2036-2047

Ward, M.H., T.M. deKok, P. Levallois, J. Brender, G. Gulis, B.T. Nolan, and J. VanDerslice. 2005. Workgroup report: drinking-water nitrate and health- recent findings and research needs. *Environmental Health Perspectives* 113(11):1607-1614.

Ward, M.H., T.M. deKok, P. Levallois, J. Brender, G. Gulis, J. VanDerslice, and B.T. Nolan. 2006. Dietary nitrate: Ward et al. respond. *Environmental Health Perspectives* 114(8): A459-A461

Wetzel, R.G. 2001. *Limnology: Lake and River Ecosystems.* 3rd ed. Academic Press, New York, NY.

Whitall, D.R. and H.W. Paerl. 2001. Spatiotemporal variability of wet atmospheric nitrogen deposition to the Neuse River Estuary, North Carolina. *Journal of Environmental Quality* 30:1508-1515.

Williams, E.J., K. Baumann, J.M. Roberts, S.B. Bertman, R.B. Norton, F.C. Fehsenfeld, S.R. Springston, L.J. Nunnermacker, L. Newman, K. Olszyna, J. Meagher, B. Hartsell, E. Edgerton, J.R. Pearson, and M.O. Rodgers. 1998. Intercomparison of ground-based NO_y measurement techniques. *Journal of Geophysical Research-Atmospheres* 103:22261-22280.

Woodman, J.N., and E.B. Cowling. 1987. Airborne chemicals and forest health. *Environmental Science and Technology* 21:120-126.

World Resources Institute. 2008. *Eutrophication and Hypoxia in Coastal Areas: a Global Assessment of the State of Knowledge.* World Resources Institute Policy Note #1, March 2008, World Resources Institute, Washington, DC

Wurtsbaugh, W.A., H.P. Gross, C. Luecke, and P. Budy. 1997. Nutrient limitation of oligotrophic sockeye salmon lakes of Idaho (USA). *Verhandlungen der Internationalen Vereinigung für Theoretische und Angewandte Limnologie* 26:413-419.

Wyers, G. P., R.P. Oties, and J. Slanina. 1993. A continuous-flow denuder for the measurement of ambient concentrations and surface-exchange fluxes of ammonia. *Atmospheric Environment* 27:2085-2090.

Xu, H.. H.W. Paerl, B. Qin, G. Zhu, and G. Gao. 2010. Nitrogen and phosphorus inputs control phytoplankton growth in eutrophic Lake Taihu, China. *Limnology and Oceanography* 55:420-432.

Yienger, J.J. and H. Levy. 1995. Empirical-Model of Global Soil-Biogenic NO_x Emissions. *Journal of Geophysical Research-Atmospheres* 100:11447-11464.

Zhang, L., A. Wiebe, R. Vet, C. Mihele, J.M. O'Brien, S. Iqbal, and Z. Liang. 2008. Measurements of reactive oxidized nitrogen at eight Canadian rural sites. *Atmospheric Environment* 42: 8065-8078.

Zhou, X.L., K. Civerolo, H.P. Dai, G. Huang, J. Schwab, and K. Demerjian. 2002. Summertime nitrous acid chemistry in the atmospheric boundary layer at a rural site in New York State, *Journal of Geophysical Research-Atmospheres* 107(D21), 4590, doi:10.1029/2001JD001539.

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http://www.epa.gov/sab
EPA-SAB-11-013
August 2011

SAB