

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 N₂, 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 (NH₃) and ammonium ion (NH₄⁺)], inorganic chemically oxidized forms of N [e.g., nitrogen oxides (NO_x), nitric acid (HNO₃), nitrous oxide (N₂O), N₂O₅, HONO, peroxy acetyl compounds such as peroxyacetyl nitrate (PAN), and nitrate ion (NO₃⁻)], 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 (N₂), 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 N₂ 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 N₂ from the atmosphere and convert it to ammonia (NH₃) – 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 losses⁷ 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 Nr⁸ 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 N₂ 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 N₂. 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

⁷ In the context of this report, “losses” refers to transfers among systems and not the conversion of Nr to N₂. Whenever N₂ 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.

The Nitrogen Cascade

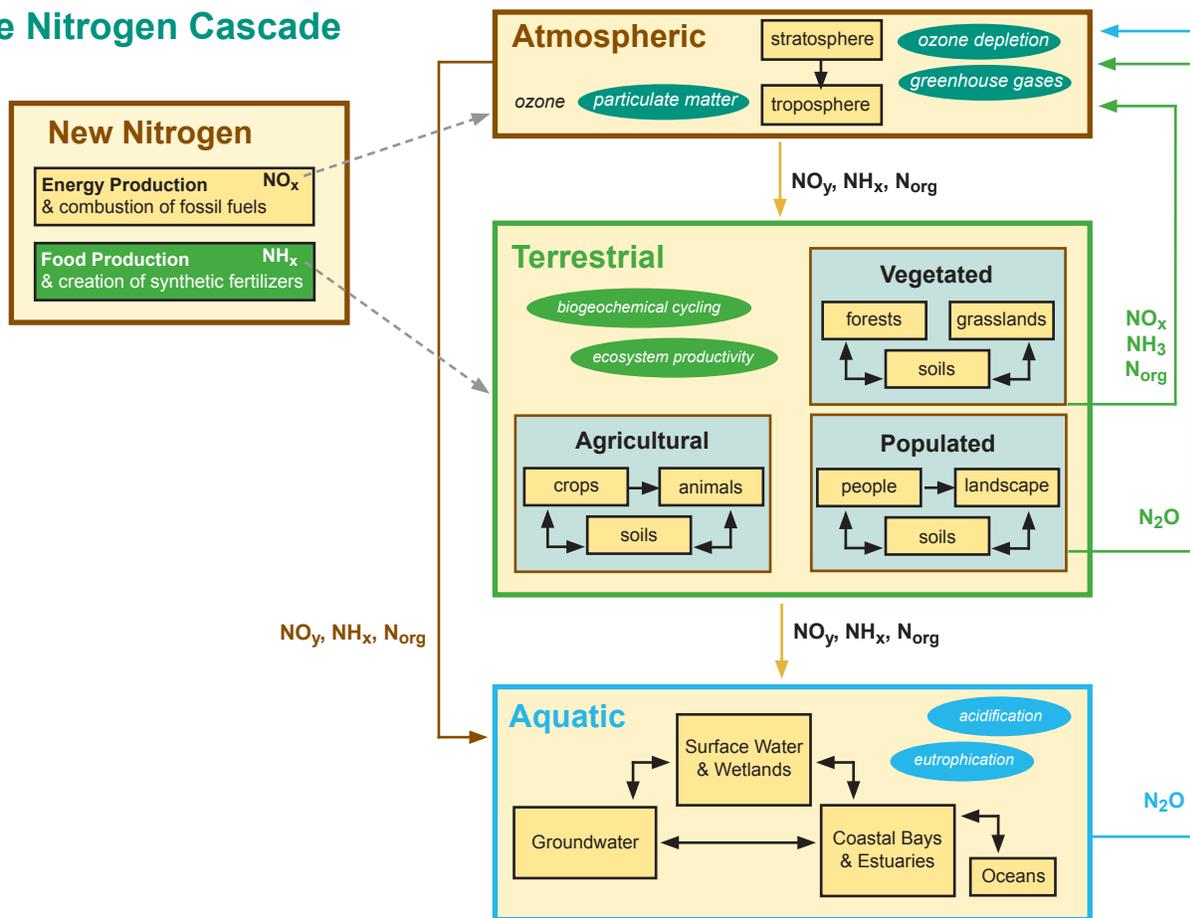


Figure 1: The nitrogen cascade

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 oxides⁹ (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 nitrogen¹⁰ (NO_y), reduced nitrogen¹¹ (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,

⁹ NO_x (oxides of nitrogen) includes $\text{NO} + \text{NO}_2$

¹⁰ NO_y (total oxidized nitrogen) includes $\text{NO}, \text{NO}_2, \text{NO}_3, \text{N}_2\text{O}_5, \text{HONO}, \text{HNO}_3, \text{NO}_3^-, \text{PAN}$ and other organo-nitrates, RONO_2

¹¹ NH_x (reduced nitrogen) includes $\text{NH}_3 + \text{NH}_4$

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 (≤ 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 strategy¹² could make to environmental protection;

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.

