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Introduction

1.1. THE SIGNIFICANCE OF NITROGEN ON EARTH

Nitrogen is essential to sustain life on Earth, as it is a major component of certain essential amino acids and proteins, enzymes, vitamins, and DNA and RNA. Also, nitrogen is contained in chlorophyll, the green pigment in plants that is essential for photosynthesis. Nitrogen, as dinitrogen (N_2) gas is the most abundant element in the Earth's atmosphere making up 78% of all gases by volume. Galloway et al. (2003) estimated that the total amount of nitrogen in the atmosphere, soils, and waters of Earth is approximately 4×10^{21} g, which is more than the combined mass of all four other essential elements (carbon, phosphorus, oxygen, and sulfur) that are needed to sustain life. However, rather ironically, this bountiful form of nitrogen (N_2 gas) is unusable by most organisms. The strong triple bond between the nitrogen atoms in nitrogen gas molecules makes N_2 gas essentially unreactive. Fortunately, certain specialized nitrogen-fixing microorganisms in soils can transform or convert nitrogen gas to forms that are available to plants (e.g., ammonium, nitrate), thereby providing sustenance to all animal life on this planet. The process by which nitrogen gas is converted by microbes into inorganic nitrogen compounds, such as ammonia, is referred to as biological nitrogen fixation (BNF). BNF accounts for approximately 90% of this transformation, which is performed by certain bacteria (e.g., genus *Rhizobium*) and blue-green algae (cyanobacteria). Small amounts of nitrogen gas fixation can occur abiotically, through high temperature processes, such as lightning and ultraviolet radiation, both of which can break the strong triple bond in molecular nitrogen (N_2) in the atmosphere. Denitrification is another important process in which microorganisms consume nitrate and convert it to reduced forms of nitrogen and ultimately back to nitrogen gas. This and other key processes of the nitrogen cycle are discussed in more detail in Chapter 2.

1.2. CYCLING OF NITROGEN IN THE ENVIRONMENT

Nitrogen and its various forms can move or cycle between the atmosphere, biosphere, and hydrosphere (Fig. 1.1). The processes involved in the transfer of the different forms of nitrogen between these systems or reservoirs within these systems are collectively referred to as the nitrogen cycle. Prior to the 19th century, the reactive nitrogen produced by BNF and abiotic processes was balanced by plant uptake and denitrification processes and therefore did not accumulate in the environment. As human population increased substantially during the 19th century, there were two main needs for reactive nitrogen: fertilizers and explosives. To meet these needs, large amounts of nitrogen were mined from various sources including naturally occurring nitrate deposits, guano, and coal. The overall dependence on the use of these sources in Europe has been referred to as a "fossil nitrogen economy" (Sutton et al., 2011a). By the end of the 19th century, these sources could not support the growing needs for more reactive nitrogen. Not long after the end of the 19th century, a solution to this problem was discovered. A process was developed in a laboratory in the early 20th century by Fritz Haber (who received a Nobel Prize for Chemistry in 1918) that synthesized ammonia (NH_3) from nitrogen and hydrogen under high temperature and pressure. This process was industrialized by Carl Bosch (who received a Noble Prize in 1931) utilizing an iron catalyst along with high temperature (300–500°C) and high pressure (20 MPa) (Fowler et al., 2013). Thus, the combined discoveries of these two men became known as the Haber–Bosch industrial chemical process, which converts hydrogen and atmospheric nitrogen gases

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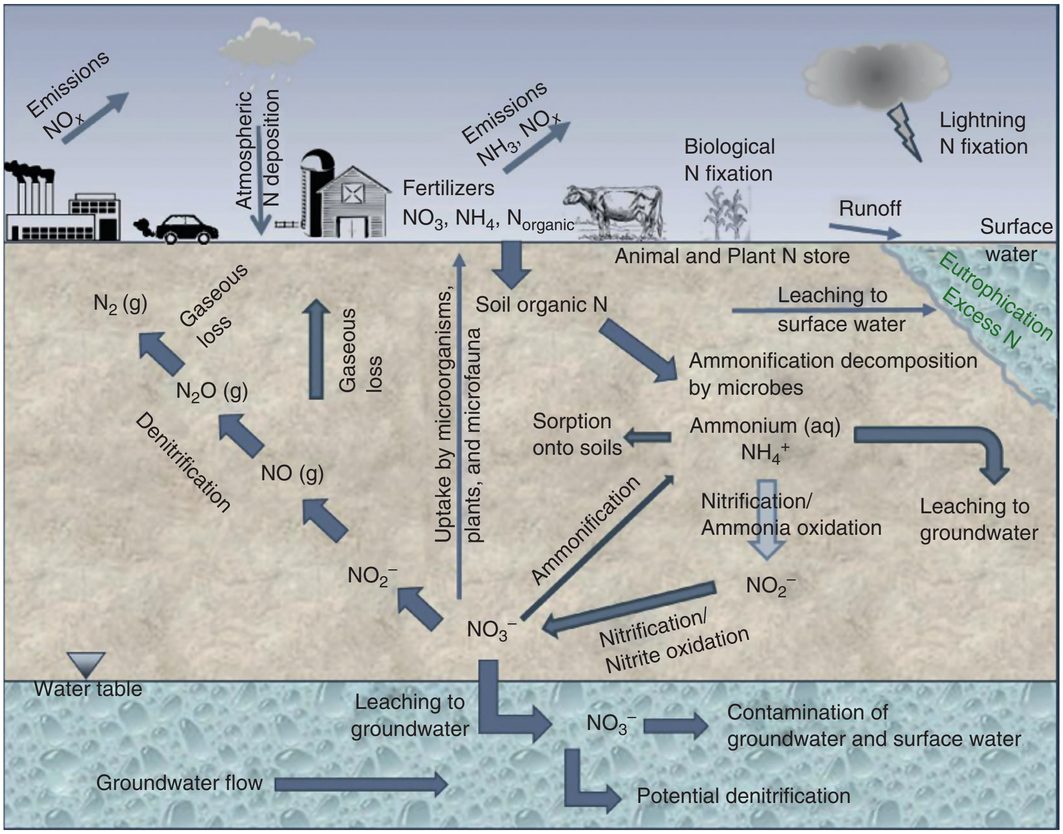

Figure 1.1 A conceptual diagram showing sources of anthropogenic and natural sources of reactive nitrogen, the various forms of nitrogen, and processes in the nitrogen cycle in the atmosphere, hydrosphere, and biosphere. *Source*: Modified from Rivett et al. (2008).

by chemical reactions into synthetic ammonia. This process is used mainly to create inorganic fertilizers, which started the extensive human alteration of the natural nitrogen cycle. An estimated 128 teragrams (Tg, 1×10^{12} g; 1 billion kg) of reactive nitrogen are synthetically produced annually from the Haber–Bosch process (Galloway et al., 2008) and used to make fertilizers for agriculture, lawns, and recreational turf grass. Sutton et al. (2011a) have referred to these large amounts of reactive nitrogen produced by the Haber–Bosch process as the "greatest single experiment in global geo-engineering that humans ever made." The production of artificial nitrogen fertilizers has grown exponentially since the 1950s and is projected to grow into the future due to increasing demand and utilization (e.g., Erisman et al., 2015; Nielsen, 2005). The abundance of inexpensive fertilizer led to its excessive use throughout the industrialized world, which created a surplus of nitrogen that was followed by substantial releases of reactive nitrogen to the environment. Global increases in reactive nitrogen have resulted from intensive cultivation of legumes, rice, and other crops that result in the conversion of nitrogen gas (N_2) to organic nitrogen compounds through human-induced BNF. The production of reactive nitrogen from Haber–Bosch process was projected to increase from 120 Tg N/yr in 2010 to about 160 Tg N/yr in 2100 (Fowler et al., 2013).

The anthropogenic production of reactive nitrogen has increased steadily and significantly since the mid-20th century as world population increased substantially (Fig. 1.2). Accompanying this increase in population are steady increases in meat and grain production, BNF, and NO_x emissions. Galloway et al. (2003) estimated that reactive nitrogen inputs resulting from cultivation of legumes and other aforementioned crops (BNF) increased globally from approximately 15 Tg N/yr in 1860 to approximately 33 Tg N/yr in 2000. To increase productivity and crop yield per acre worldwide, increased amounts of fertilizer were being using along with fossil fuel burning machines that replace practices that involved manpower and the use of farm animals (Fig. 1.2). Throughout Europe, regional watersheds annually contribute approximately 3700 kg of reactive nitrogen per square kilometer, which is five times the background rate of natural N_2 fixation (Sutton et al., 2011a). On a global scale, as a result of intensive farming practices, nitrogen and other nutrients were and are being depleted in some areas and have been concentrated in

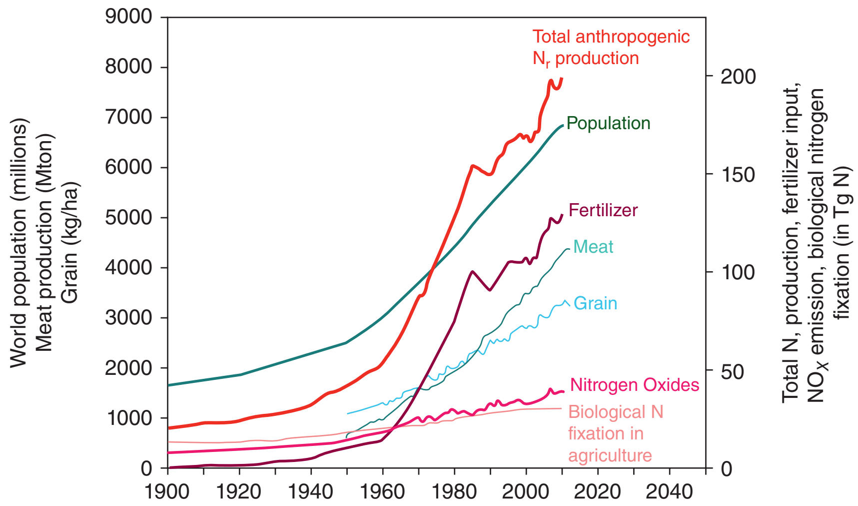

Figure 1.2 Plot showing increases in human population and increases in the creation of total anthropogenic reactive nitrogen between 1900 and 2012 (Mton, is millions of tons; kg/ha is kilograms per hectare; and Tg N is teragrams (10^{12} g) of reactive nitrogen). *Source*: With permission from Erisman et al. (2015). Reproduced with permission of WWF.

other areas, leading to a "cascade" of reactive nitrogen (Galloway et al., 2003) through the environment and creating a sequence of harmful environmental effects including ecosystem damages (loss of biodiversity, eutrophication of waters and soils, and soil acidification), increases in greenhouse gas emissions, contamination of drinking water, air pollution, human health maladies, and damages to the ozone layer (Galloway et al., 2008).

In addition to inputs of reactive nitrogen from the use of synthetic fertilizers, there are many other point and nonpoint anthropogenic sources that release reactive nitrogen to the environment (Fig. 1.1). Additional amounts of reactive nitrogen are contributed from fossil fuel combustion, which converts both atmospheric nitrogen and fossil nitrogen to reactive oxidized forms (NO_x). The amount of reactive nitrogen created from fossil fuel combustion increased from less than 1 Tg N/yr in 1860 to approximately 25 Tg N/yr in 2000 (Galloway et al., 2003; Nielsen, 2005). Anthropogenic inputs of nitrogen to the land surface from animal wastes, human wastewater (septic tanks, wastewater treatment facilities), industrial processes, and atmospheric deposition have also contributed to the substantial alteration of the cycling of nitrogen throughout the world.

The overall fate of anthropogenic reactive nitrogen in the terrestrial biosphere is not well understood. As Galloway et al. (2008) noted, the fate of only 35% of the reactive nitrogen inputs to the terrestrial biosphere were known, which included estimates of 18% exported to and denitrified in coastal ecosystems, 13% deposited to the ocean from the marine atmosphere, and 4% emitted as N_2O. Although tropical forests cover only 12% of the earth's surface, it is estimated that they emit about 50% of the N_2O to the atmosphere (Townsend et al., 2011). The approximately 65% of the remaining anthropogenic reactive nitrogen is potentially accumulated in soils, vegetation, and groundwater or possibly denitrified to nitrogen gas. However, these estimates have a considerable amount of uncertainty (Galloway et al., 2008).

Overall, freshwater nitrogen loadings worldwide have increased from 21 million tons in preindustrial times to 40 million tons per year in 2009 (Dodds et al., 2009), while riverine transport of dissolved inorganic nitrogen has increased from 2–3 million to 15 million tons. During the last part of the 20th century, increased nitrogen loading to the land surface has caused the natural rate of N fixation to double and the atmospheric deposition rates to increase more than tenfold. This so-called "greatest single experiment in global geo-engineering that human ever made" (Sutton et al., 2011a) has led to a legacy of detrimental consequences for human health, terrestrial and coastal ecosystems, and the economy. We now explore in more detail some of these adverse consequences from excess reactive nitrogen in our environment.

1.3. ENVIRONMENTAL CONSEQUENCES OF EXCESS REACTIVE NITROGEN

During the past 50 years, excess reactive nitrogen has had dire consequences for many varied types of environmental systems all over the world (Erisman et al., 2011, 2013, 2015; Payne et al., 2013; Townsend et al., 2003; B. Ward, 2012). As mentioned previously, only a small proportion of the Earth's biota can transform N_2 to reactive nitrogen forms. As a result, reactive nitrogen tends to be the limiting nutrient in most natural ecosystems and

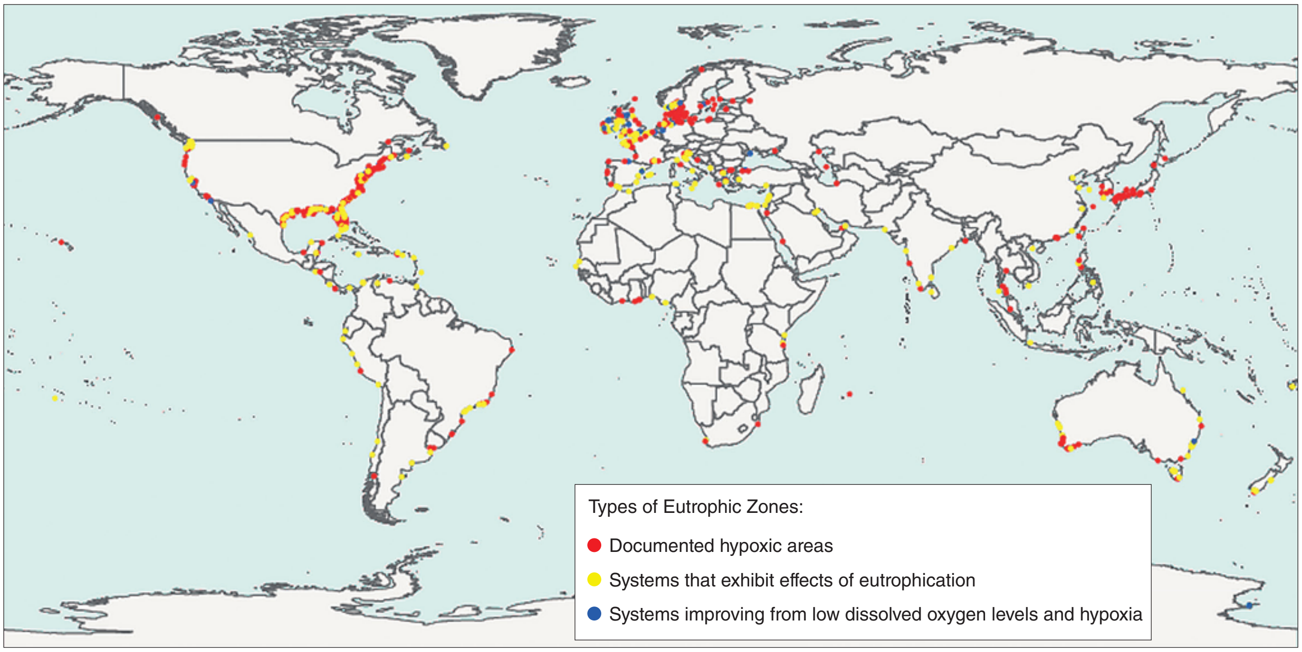

Figure 1.3 Map showing hypoxic (red circles) and eutrophic (yellow circles) coastal areas around the world. *Source*: Modified from the colored figure 4 in Erisman et al. (2015). Reproduced with permission of World Wildlife Fund.

typically for agricultural systems. Therefore, increased amounts of reactive nitrogen in the environment have led to shifts in plant species composition, decreases in species diversity, habitat degradation, eutrophication, decreases in water transparency, increased biomass of benthic and epiphytic algae, and changes in food-web processes in terrestrial ecosystems and in coastal waters (Howarth et al., 2000, 2011). These effects are more prevalent in certain areas, such as the highly affected areas in Central and Western Europe, Southern Asia, the eastern United States, and parts of Africa and South America (Erisman et al., 2015).

Excess nitrogen in water bodies have led to the proliferation of harmful (toxic) algal blooms, which have been reported in every state in the United States during the past 10 years (U.S. Environmental Protection Agency (U.S. EPA), 2009), as well in as many other parts of the world (Erisman et al., 2015). Nonaquatic animals also are affected when consuming waters-containing algae. Pets and livestock have died after drinking water containing algal blooms, including 32 cattle on an Oregon ranch in July 2017 (Flesher & Kastanis, 2017). Algal blooms have resulted in numerous beach closures in Florida and a state of emergency was declared in four coastal counties in southeast Florida in 2016. Toxic algal blooms have contaminated waterways from the Great Lakes to Chesapeake Bay, from the Snake River in Idaho to New York's Finger Lakes and reservoirs in California's Central Valley (Flesher & Kastanis, 2017). As of mid-August 2016, the U.S. EPA noted that states across the United States have reported more than 250 health advisories due to harmful algal blooms that year. The National Aquatic Resource Surveys conducted by the U.S. EPA and state and tribal partners in 2012 found that 34% of the lakes surveyed in the United States had high levels of nitrogen associated with harmful ecological impacts. A survey of rivers and streams in 2009–2010 found that 41% had high levels of nitrogen (https://www.epa.gov/national-aquatic-resource-surveys/data-national-aquatic-resource-surveys, accessed 19 October 2018). Large algal blooms occurred in the Black Sea in the 1970s and 1980s following the intensive use of fertilizers and livestock production in the Black Sea basin in the 1960s (Bodeanu, 1993; Mee et al., 2005). Most of Europe has a high potential risk of eutrophication of surface freshwaters (Grizzetti et al., 2011) as well as parts of the United States, South America, Australia, and Southeast Asia (Erisman et al., 2015) (Fig. 1.3).

Another severe impact of excessive amounts of reactive nitrogen is the depletion of oxygen (hypoxia), which has led to dead zones in estuaries and other large water bodies. In the United States more than 166 dead zones have been identified and have affected waterbodies such as the Chesapeake Bay (http://www.cbf.org/issues/dead-zones/, accessed 18 October 2018) and the Gulf of Mexico (Rabalais et al., 2002). The Gulf of Mexico dead zone grew to approximately 5840 $mile^2$ in 2013. This extremely large dead zone results from summertime nutrient pollution from the Mississippi River (excess reactive nitrogen most likely from fertilizers), which drains 31 upstream states (https://www.epa.gov/nutrientpollution/

sources-and-solutions). Recent studies have indicated that there could be more than 500 coastal dead zones worldwide (Fig. 1.3), with numbers possibly doubling each decade (Breitburg et al., 2018, Conley et al., 2009, Diaz & Rosenberg, 2008). Dead zones have increased in large coastal areas of the Adriatic Sea, the Black Sea, the Kattegat, and the Baltic Sea (Diaz & Rosenberg, 2008).

Nitrogen gases, aerosols, and dissolved compounds in air that contain NO, N_2O, nitrate, ammonia, and ammonium can have injurious and phytotoxic effects on the above ground parts of plants (e.g., trees, shrubs, and other vegetation). In the 1980s, foliar impacts to forests were prevalent in North America and Europe due to air pollution containing nitric and sulfuric acids; however, the situation has improved in these areas with air-pollution control legislation and pollution reduction strategies adopted by some industries. Increased amounts of nitrogen oxides in the atmosphere could also contribute to global climate change and stratospheric ozone depletion (Galloway et al., 2004). Ironically, one potential benefit from increased nitrous oxide emissions is stimulating the sequestration of global CO_2 by terrestrial and marine ecosystems. This effect could possibly lower the amounts of the greenhouse gas CO_2 released to the atmosphere (Fowler et al., 2015; Zaehle et al., 2011).

Atmospheric deposition of reactive nitrogen also leads to acidification of soils and surface waters. During 2000–2010, Peñuelas et al. (2013) reported that global land had received more than 50 kg/ha accumulated N deposition. They found that nitrogen additions to the land surface significantly reduced soil pH by 0.26 on average globally. Soils in many places around the world are particularly sensitive to nitrogen deposition, with buffering likely transitioning from base cations (Ca, Mg, and K) to nonbase cations (Al and Mn), which could have a toxic impact for terrestrial ecosystems (Bowman et al., 2008; Tian & Niu, 2015). Significant decreases in forest and grassland productivity have been observed when reactive nitrogen deposition increases above threshold levels. These adverse effects could promote biodiversity changes through the food chain, affecting insects, birds, and other animals that depend on these food sources (Erisman et al., 2013). Acidification of surface waters (lakes, streams, oceans) in many places around the world also has increased and has had detrimental effects on biota (Curtis et al., 2005; Doney et al., 2007).

Ammonia emissions to the atmosphere also have increased during the past several decades (Ackerman et al., 2018) and can result in the formation of fine particulate matter through reactions with nitric and sulfuric acids. These harmful compounds in the atmosphere can be dispersed over large areas of the world, resulting in eutrophication and acidification of terrestrial, freshwater, and marine habitats (Bobbink et al., 2010; Erisman et al., 2015). Also, deposition of ammonia can damage sensitive vegetation, particularly bryophytes and lichens that consume most of their nutrients from the atmosphere (Sheppard et al., 2011). Sala et al. (2000) noted that certain biomes (particularly northern temperate, boreal, arctic, alpine, grassland, savannah, and Mediterranean) are very sensitive to reactive nitrogen deposition because of the limited availability of nitrogen in these systems under natural conditions.

1.4. ADVERSE HUMAN HEALTH IMPACTS

The widespread alteration of the nitrogen cycle from the production and use of synthetic fertilizers has both positive and negative consequences for human health. One cannot discount the huge benefits from nitrogen fertilizer use in developing countries, which has led to an increase in food production and substantial reductions in malnutrition. With better nutrition, healthier diets can lead to more efficient immune response to parasitic and infectious diseases (Nesheim, 1993). Although, shortages of food and malnutrition still exist in many large parts of the world.

Increases in reactive nitrogen compounds in the atmosphere (air pollution) and drinking water have had direct negative effects on human health. Townsend et al. (2003) show schematically how the net public health benefit from human fixation and use of reactive nitrogen has peaked and declined as a result of an exponential increase in air and water pollution and ecological feedbacks to disease (Fig. 1.4). Increased emissions of nitrogen oxides in urban areas have resulted from fossil fuel combustion. In other areas, biomass burning and volatilization of fertilizers emit large quantities of reactive

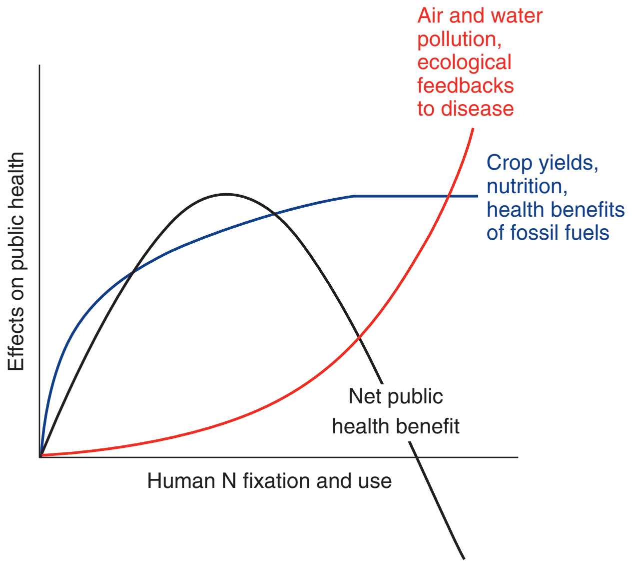

Figure 1.4 Conceptual diagram showing detrimental effects on human health from increasing amounts of excess reactive nitrogen from human fixation. *Source*: From Townsend et al. (2003). Reproduced with permission of John Wiley & Sons.

nitrogen, including ammonia and nitrogen oxides. Elevated nitrogen oxides enhance the production of tropospheric ozone (O_3) at low altitudes and aerosols, which can cause coughs, asthma, and other reactive airways disease, and mortality (Apte et al., 2015; Townsend et al., 2003). At high altitudes and in parts of the stratosphere, ozone can be destroyed due to the production of the catalyst NO as ultraviolet light breaks N_2O apart. The destruction of ozone in the stratosphere would allow more ultraviolet light to reach the Earth's surface, possibly causing more skin cancers. Kane (1998) noted that reductions in ozone may result in a 10–20% increase in ultraviolet-B radiation, and this could explain a 20–40% rise in skin cancer in the human population since the 1970s. Particulate air pollution has also been linked to cardiovascular diseases and overall mortality (Pope et al., 2002). N_2O is a potent greenhouse gas with a long residence time in the atmosphere of 120 years (Howarth et al., 2011) and therefore its harmful effects can persist for decades.

Humans are increasingly exposed to elevated levels of reactive nitrogen species (mainly nitrate) in drinking water. The U.S. EPA has set a maximum contaminant level of 10 mg/L (parts per million) for nitrate-N in drinking water. In the United States, it is estimated that 10–20% of groundwater sources may exceed 10 mg/L for nitrate-N (Dubrovsky et al., 2010). In 2015, the U.S. EPA reported that 183 community water systems exceeded allowable levels of nitrate in drinking water. Sutton et al. (2011a) estimated that about 3% of the population in 15 European Union countries (EU-15) that relies on groundwater as a drinking water source is exposed to nitrate concentrations exceeding the drinking water standard of 50 mg NO_3/L (equivalent to 11.2 mg N/L, expressed as nitrogen). In addition, it was estimated that 5% of the population using groundwater is chronically exposed to nitrate concentrations exceeding 5.6 mgN/L, which could double the risk of colon cancer for people that consume above median amounts of meat (Sutton et al., 2011a). In many other aquifers around the world, nitrate-N concentrations exceed the maximum contaminant level set by the World Health Organization. Many large aquifers in China have been contaminated with elevated nitrate-N concentrations (S. Wang et al., 2016b). About 90% of China's shallow groundwater is polluted, and nitrate is considered one of the pollutants of main concern (Qiu, 2011).

Numerous health effects have been associated with elevated nitrate levels in drinking water, including methemoglobinemia (elevated levels of methemoglobin in blood which leads to decreased oxygen availability to tissues, mostly affects babies), reproductive problems, and cancer. Recent studies during the past 20 years have reported that nitrate-N concentrations even below the maximum contaminant level of 10 mg/L can cause other health maladies including non-Hodgkins's lymphoma (M.H. Ward et al., 1996, 2005, 2018) and increased risk of bladder and ovarian cancers (Weyer et al., 2001).

A developing area of interdisciplinary research is investigating the links between ecosystem damages and human diseases. For example, Townsend et al. (2003) hypothesized that an increase in nutrient availability can favor opportunistic, disease-causing organisms and could lead to changes in the epidemiology of human diseases. Johnson et al. (2010) referred to studies that showed that ecological changes related to nutrient enrichment often aggravate infection and disease caused by "generalist" parasites with direct or simple life cycles. More recently, reports of attacks to the immune systems of sport fisherman in Lake Erie were linked to algal blooms (Flesher & Kastanis, 2017). Studies have reported associations between algal blooms with cholera outbreaks and correlations between inorganic nutrient concentrations and abundances of mosquitoes that can carry pathogenic microorganisms (Camargo & Alonso, 2006).

1.5. LEGACY REACTIVE NITROGEN STORAGE IN THE SUBSURFACE

Comparably disturbing, but not widely acknowledged, are the large amounts of reactive nitrogen, mostly in the form of nitrate, that leaches below the root zone in soils to the vadose (unsaturated) zone (the material between the base of the soil to the water table at the top of the saturated zone) and eventually to groundwater (aquifers) where nitrate can be stored (Walvoord et al., 2003; L. Wang et al., 2013, 2016a). This is especially prevalent in agricultural areas where there has been a surplus of nitrogen added to cropland. Ascott et al. (2017) assessed global patterns of nitrate storage in the vadose zone by using estimates of groundwater depth and nitrate leaching for the period 1900–2000. They estimated a peak global storage of 605–1814 Tg of nitrate in the vadose zone, with the highest storage per unit area in North America, China, and Europe (areas with thick vadose zones and extensive historical agricultural activities).

Several recent studies have shown that groundwater nitrogen applied decades ago can still be found in aquifers (Basu et al., 2012; McMahon et al., 2006; Meals et al., 2010; Puckett et al., 2011; Sanford & Pope, 2013; Sebillo et al., 2013). Legacy nitrogen stored in the subsurface over long periods of time can slowly be released to streams and large rivers (Van Meter & Basu, 2015, 2017; Van Meter et al., 2016, 2017). However, even though substantial decreases in fertilizer applications have been noted in the Netherlands, Denmark, and Germany (where the nitrogen surplus is back to the level of that of 1970), concentrations of nitrate-N in groundwater have not responded to this decrease of nitrogen surplus (Sutton et al., 2011a). Particularly noteworthy are cases in Eastern European countries where the nitrogen surplus has decreased by half (due to economic and political changes in the early 1990s), however, no improvements in water quality have been observed in streams (Sutton et al., 2011a). This is likely due to the large quantities of nitrate stored in aquifers and released very slowly over time, as a function of the groundwater residence time, which can range from weeks to several thousands of years (Alley et al., 2002; Tesoriero et al., 2013).

In arid environmental systems, an increased reliance on dryland ecosystems for agriculture may result in flushing of naturally occurring nitrate in the unsaturated zone into groundwater in regional basins due to irrigation and vegetation change (Robertson et al., 2017). Thus, the unsaturated zone, an aforementioned important reservoir for the storage and release of reactive nitrogen (mainly in the form of nitrate) over time, has significant implications for global warming, contamination of deeper aquifers used for drinking water supplies, and eutrophication of surface water bodies (Schlesinger, 2009).

1.6. ECONOMIC IMPACTS OF EXCESS REACTIVE NITROGEN

Environmental degradation from excess reactive nitrogen has led to severe economic consequences, mainly associated with treating drinking water and wastewaters, and human health issues associated with air pollution and surface water and groundwater contamination. Human-caused eutrophication of waterways from excess nutrients has resulted in large revenue losses to the commercial fishing industry (aquaculture, fisheries, and shellfish fisheries), tourism and recreational water usage, loss of real estate values for waterfront property, and threatened and endangered species. Dodds et al. (2009) provided a detailed economic analysis of the losses associated with environmental impacts related to eutrophication in the United States and reported a conservative estimate of potential losses totaling over \$2.2 billion annually. This estimate did not include additional water treatment costs related to taste and odor problems. Sobota et al. (2015) estimated reactive nitrogen leakage for large watersheds in the United States and accounted for economic costs associated with mitigation, remediation, direct damage, human health, agriculture, ecosystems, and climate. They reported estimated annual damage costs of up to \$2255/ha across watersheds, with a median value of \$252/ha/yr. For the entire United States, Sobota et al. (2015) estimated environmental damages from reactive nitrogen in the early 2000s that ranged from \$81 to \$441 billion per year.

Based on information from multiple studies conducted during 2000 through 2012, the U.S. EPA compiled extensive information on costs associated with adverse impacts of nutrient pollution in the environment and reducing nutrient pollution from various point and nonpoint sources (U.S. EPA, 2015). The following categories were identified in the report and demonstrate how costs can have cascading economic impacts through many different socioeconomic sectors. For example, losses in tourism and recreation associated with algal blooms can include decreased restaurant sales and hotel stays, loss of tourism-related jobs, closure of lakeside businesses, and decreased tourism-related spending. Negative impacts to commercial fishing throughout coastal areas has included reduced harvests, fishery closures, loss of jobs, and increased processing costs related to elevated risks associated with shellfish poisoning. There also have been significant losses in property values of waterfront property and nearby homes associated with decreases in water clarity, algal mats, and increased pollutant concentrations. Algal blooms have caused a variety of injurious health effects in humans and animals through direct contact with skin during recreational activities, drinking water consumption, intake of contaminated shellfish (can cause neurotoxic shellfish poisoning and other serious health effects). A study in Florida, reported that increased emergency room visits cost Sarasota County more than \$130,000 (in 2012 dollars) to treat respiratory illnesses associated with increased algal blooms (Hoagland et al., 2009). Waters containing algal toxins and with taste and odor problems have significant treatment costs. The U.S. EPA 2015 report also estimated the extensive mitigation and restoration costs for coastal and inland waterways contaminated with algal blooms. Also, various organic forms of nitrogen can be stored in bottom sediments of lakes, estuaries, and other waterbodies and then be released to the water column during storms and other weather-related events. In-lake treatment and mitigation costs may include aeration, alum treatments, biomanipulation, and dredging. There are enormous costs to Federal, State, and local agencies associated with restoring impaired waterbodies, including developing watershed management plans, total maximum daily loads (TMDLs), nutrient trading and other programs. High costs associated with nutrient pollution control from point and nonpoint sources include upgrades to municipal wastewater treatment plants (advanced wastewater treatment to meet TMDLs for impaired waters), conversion of septic tanks to centralized wastewater treatment systems, industrial and agricultural wastewater treatment, and the control of elevated levels of nitrogen in stormwater runoff.

Over the past two decades, the State of Florida has spent hundreds of millions of dollars on restoration of large springs and their spring runs with impaired water quality and adversely affected ecosystems. More than 1000 springs have been identified in Florida, and over the past several decades, their water quality has become degraded (Katz, 2004; Scott, 2002). Elevated nitrate-nitrogen concentrations have resulted in excessive algal growth that has depleted aquatic oxygen levels and negatively impacted the health of the spring ecosystem. About 30 of these springs have been given special status because they have been deemed by the Florida Legislature to be a unique part of the state's scenic beauty, provide critical habitat, and have immeasurable natural, recreational, economic, and inherent value. Studies have indicated that springs contribute significantly to Florida's economy, with more than 100 million dollars per year from visitor spending, jobs, and associated hotel stays and sales at restaurants, dive shops, and other expenses. In 2016, the Florida Legislature enacted the "Florida Springs and Aquifer Protection Act," which provides continued funding ($50 million per year) and special status and protection to historic first-magnitude springs (each discharging more than 2.8 m^3/s of water from the Floridan aquifer) and to other springs of special significance. The act requires the Florida Department of Environmental Protection to set restoration targets and adopt a basin management action plan for each water-quality impaired system to reduce nutrient loading and improve water quality over the next 20 years.

In England and Wales, Pretty et al. (2003) estimated the damage costs of freshwater eutrophication in to be $105–160 million per year (£75.0 million to 114.3 million). The damage costs were grouped into seven categories, each with costs of $15 million per year or more: (a) reduced value of waterfront dwellings, (b) drinking water treatment costs for nitrogen removal, (c) reduced recreational and aesthetic value of water bodies, (d) drinking water treatment costs for removing of algal toxins and decomposition products, (e) production of greenhouse and acidifying gases, (f) negative ecological effects on biota, and (g) net economic losses from the tourist industry. In China, Qiu (2011) estimates that billions of dollars will be spent cleaning up contaminated groundwater related to excess nutrients.

The European Nitrogen Assessment (ENA) program (Brink et al., 2011) estimated that the total annual environmental damage related to reactive nitrogen ranged between 70 and 320 billion euro (in terms of 2009 euro), which was equivalent to 150–175 euro per capita. About 75% of this estimate was attributed to health damage and air pollution. This constituted 1–4% of the average European income. The studies also reported the following six societal costs per kilogram of reactive nitrogen in pollutant emission (in euro/kg N): (a) health impacts (10–30), (b) health effects from secondary ammonium particles (2–20), (c) greenhouse gas balance effects of N_2O (5–15), (d) ecosystem impacts from nitrogen runoff (5–20), (e) nitrogen deposition (2–10), and (f) N_2O associated with stratospheric ozone depletion (1–3). Preliminary estimates of global damage costs associated with nitrogen pollution range from 200 to 2000 billion USD per year (Sutton et al., 2013).

Numerous studies have documented the enormous costs associated with the treatment of drinking water in the United States (to remove algal toxins and algal decomposition products) and advanced treatment of human wastewaters (municipal wastewater treatment facilities) and management practices to process animal wastes from confined animal feeding operations. Collectively, federal and state agencies have spent billions of dollars on trying to alleviate water-quality degradation from excess nitrogen; however, recent studies have shown little to no improvement in water quality. In 2004, the Drinking Water Infrastructure Needs Survey estimated that $150.9 billion will need to be spent on drinking water systems to provide safe treatment, storage, and distribution (U.S. EPA, 2004). A 2009 report by the State-U.S. EPA Nutrient Innovations Task Group (2009) noted that current nutrient control strategies are woefully inadequate and the rate of nutrient pollution will likely increase as the world population continues to increase. The population of the United States is projected to increase by more than 135 million over the next 40 years. As population grows, more nutrients (mainly reactive nitrogen compounds) will be released to the environment from municipal wastewater discharges and from other sources such as wastes from agricultural livestock and row-crop runoff are likely to grow.

1.7. PURPOSE AND SCOPE

Unfortunately, many people (including students of all ages, policymakers, teachers, environmental, and economic strategists) are not aware of the extensive ecological, human health, and economic consequences of excess amounts of reactive nitrogen in the environment. This is despite the fact that over the past 40 years, there have been numerous national and international scientific studies that have generated hundreds of published reports and articles on various aspects of environmental degradation, adverse human health effects, and the enormous economic costs related to the anthropogenic alteration of the nitrogen cycle with an overload of reactive nitrogen species. For more than 25 years, Galloway et al. (1995, 2003, 2004, 2008) have quantified global inputs of reactive nitrogen and documented the consequences of their cascading effects in the atmosphere, terrestrial ecosystems, freshwater and marine systems, and human health impacts. In Europe, there have been numerous studies that provide an historical foundation on the alteration of the nitrogen cycle. During the 1970s and 1980s, the sources and impacts of nitrogen were studied throughout Europe, including assessments by the Scientific Committee on Problems of the Environment (SCOPE) (Blackburn & Sorenson, 1988; Soderlund & Svensson, 1976) and by the Royal Society of United Kingdom (Stewart & Rosswall, 1982). Mosier et al. (2004) presented an evaluation of the alteration of the nitrogen cycle in agriculture.

Undeniably, one of the most comprehensive studies of the anthropogenic alterations to the nitrogen cycle and their consequences of excess reactive nitrogen was conducted by the ENA program (Sutton et al., 2011b). This 4-year program (2007–2011) presented the first detailed continental-scale (Europe) assessment of reactive nitrogen in the environment and provided a multidisciplinary overview of the key processes in the nitrogen cycle, issues associated with up-scaling from field, farm, and city to national and continental scales, and opportunities for more effective management at local and global levels. The ENA program also was designed to inform policymakers at local and global scales by providing an extensive review of the current scientific understanding of nitrogen sources in Europe, their effects and interactions, and analyses of current policies along with associated economic costs and benefits. In addition, detailed information was presented on current nitrogen challenges in Europe and for a global context, what is known about (a) nitrogen processing in the biosphere and associated uncertainties, (b) nitrogen flows and fate at various spatial scales, (c) managing nitrogen by identifying major societal threats, and (d) strategies for more effective nitrogen management. A more recent assessment and analysis of the global nitrogen cycle with a focus on terrestrial ecosystems and the atmosphere was published by the Royal Society of the United Kingdom (Fowler et al., 2013).

As one can clearly see, the magnitude and scope of the problems associated with excess reactive nitrogen in our environment are enormous. Even with this barrage of scientific information over the past two decades that effectively demonstrates the adverse environmental, human health, and economic impacts, the message about reactive nitrogen overuse and atmospheric emissions is not making any significant impact on a wide audience. Our society must become increasingly vigilant in our commitment to address the challenges associated with significantly reducing inputs of reactive nitrogen to prevent further degradation of our surface waters, aquifers, and ecosystems. More continued collaboration and partnerships and policies are needed to develop more effective means of reducing reactive nitrogen inputs to the land and our waterways.

There is an urgent need to continue to integrate information from various disciplines on the consequences of excess reactive nitrogen in our environment and effectively communicate this information in a more coherent way so that our society can better understand these problems and move forward to develop more effective strategies to reduce environmental, human health, and economic impacts from excess nitrogen in surface waters, groundwater, and the atmosphere. Recently, there has been increased attention to understanding the footprint of reactive nitrogen generated by the consumption of nitrogen in different areas around the world. Several nitrogen footprint tools have been used to educate people and institutions where people work to support policy changes and help people make better decisions regarding resource use that have contributed to the many problems involving reactive nitrogen in the environment (Galloway et al., 2014). The importance of emphasizing the value of clean water, our most precious resource, to our society cannot be overstated.

The material presented in this book addresses this urgent and critical need for integrating and synthesizing previous and new sources of information on environmental, human health, and economic consequences of excess reactive nitrogen in our environment. The book begins with the first three chapters that present background information. This first chapter (Chapter 1) presents a concise overview of the various issues and the environmental, health, and economic consequences associated with excess reactive nitrogen. The purpose and scope of the book are also described in this chapter. Chapter 2 presents an overview of the nitrogen cycle and how the cycle has been dramatically altered during the past 100 years by anthropogenic activities. Chapter 3 provides detailed information on the major point and nonpoint sources that contribute reactive nitrogen to the environment at large regional and global scales. Also discussed are factors that affect nitrogen transport to the environment at large regional and global scales. Chapter 4 builds on this introductory information and presents information on novel and innovative tools used to identify and quantify sources of nitrate contamination. The information presented in the first four chapters is provided to help the reader gain a better understanding of reactive nitrogen cycling. The next several chapters are devoted to ramifications (and detrimental impacts) from the substantial anthropogenic alteration of the nitrogen cycle including adverse human health impacts (Chapter 5); degradation and contamination of ecosystems, surface waters, groundwater, and springs, (Chapters 6–8). Chapter 9 discusses other contaminants that co-occur with nitrate-nitrogen contamination resulting from various anthropogenic sources. Chapter 10 presents detailed information on economic consequences and costs associated with environmental pollution (e.g., human health maladies, clean-up costs, drinking-water treatment costs, losses to commercial fishing, tourism, recreational activities, decreased real estate values, and cascading effects to other socioeconomic sectors). The final chapter

(Chapter 11) discusses effective ways to reduce our nitrogen footprint (locally and globally) from human activities and presents practical strategies, and recommendations for restoring water quality in impacted surface water bodies, groundwater and springs, and terrestrial and aquatic ecosystems. Three areas are highlighted where ongoing efforts are being focused that include increasing nitrogen use efficiency and sustainability in agricultural systems, reducing the per capita consumption of animal proteins, and decreasing fossil fuel combustion and replacing with alternative energy sources. In recent years, many countries have been making important strides in identifying and communicating societal threats, and in developing strategies for integrated approaches for tackling excess reactive nitrogen in our environment.

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