

# Reactive Nitrogen and The World: 200 Years of Change

This paper examines the impact of food and energy production on the global N cycle by contrasting N flows in the late-19<sup>th</sup> century with those of the late-20<sup>th</sup> century. We have a good understanding of the amounts of reactive N created by humans, and the primary points of loss to the environment. However, we have a poor understanding of nitrogen's rate of accumulation in environmental reservoirs, which is problematic because of the cascading effects of accumulated N in the environment. The substantial regional variability in reactive nitrogen creation, its degree of distribution, and the likelihood of increased rates of reactive-N formation (especially in Asia) in the future creates a situation that calls for the development of a Total Reactive Nitrogen Approach that will optimize food and energy production and protect environmental systems.

## INTRODUCTION

The late-18<sup>th</sup> century—N discovered.

The late-19<sup>th</sup> century—natural N fixation discovered.

The late-20<sup>th</sup> century—anthropogenic N fixation exceeded natural terrestrial N fixation.

Nitrogen is the very stuff of life. It constitutes a major part of the nucleic acids that determine the genetic character of all living things and the enzyme proteins that drive the metabolic machinery of every living cell. Triple-bonded nitrogen gas (N<sub>2</sub>) makes up nearly 80% of the total mass of the Earth's atmosphere. To most organisms, this huge reservoir of N is not biologically available. Before N can be used by most plants, animals, insects, and microorganisms, the triple bonds of N<sub>2</sub> molecules must be broken and the resulting single-N atoms must be bonded chemically with one or more of 3 other essential nutrient elements—oxygen and/or hydrogen through N-fixation processes and carbon through N-assimilation processes (1).

Breaking the triple bonds within the N<sub>2</sub> molecule is an energy-requiring reaction. In nature, fixation of N<sub>2</sub> is accomplished mainly by certain unique microorganisms that have developed the special metabolic machinery necessary to produce biologically active reduced forms of nitrogen such as ammonia, amines, and amino acids—the structural constituents of proteins and nucleic acids (2). These specialized organisms include a few free-living bacteria and blue-green algae, and also certain symbiotic bacteria that have developed special metabolic relationships with the roots of leguminous crop plants such as soybeans, clover, and N-fixing trees such as alder. Oxidative fixation of gaseous N<sub>2</sub> leads to production of reactive oxidized forms of N. Oxidative fixation of N also takes place in nature, but only in high-temperature natural processes such as lightning.

In the pre-human world, biological nitrogen fixation was the dominant means by which new reactive nitrogen (Nr) (3) was made available to living organisms. The total amount of Nr that circulated naturally among various compartments of the atmosphere and the biosphere of the Earth was quite small. Thus, the biodiversity and intricate webs of relationships found in nature evolved as a result of intensive competition among many different life forms—most of them evolving under N-limited conditions.

During the last 2 centuries, human involvement with N began with discovery of N as an element (late-18<sup>th</sup> century). This led to the discovery of biological nitrogen fixation (late-19<sup>th</sup> cen-

tury), and eventually to the human dominance of the global rate of N fixation (late-20<sup>th</sup> century). This paper presents an assessment of the impact of humans on the biogeochemical cycle of N due to the enhanced creation of reactive-N. Following a historical overview, this paper discusses the changes in N-mobilization rates due to human activities, the resultant changes in the distribution fluxes to waters and the atmosphere, and an assessment of the environmental consequences of these changes. The paper concludes with an overview of regional differences in human alteration of the N cycle, casts an eye toward the future and presents the case for developing a *Total Nr Approach* for management of N.

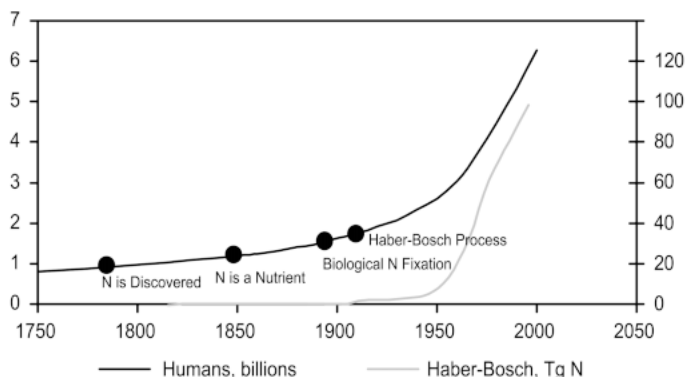
## HISTORICAL CONTEXT

Two hundred years span the time from discovery of N as an element to the present time of human dominance in Nr creation. Over that period, the global human population increased from ~ 0.85 billion to ~ 6.2 billion. This section presents significant discoveries regarding N in the context of a growing human population (Fig. 1). Unless otherwise noted, historical information is taken from Smil (4).

Nitrogen was discovered in the late 18<sup>th</sup> century through the work of several early chemists—Scheele (1742–1786, Sweden), Rutherford (1749–1819, Scotland), Lavoisier (1743–1794, France). It was formally named 'nitrogene', by the French scientist, Chaptal (1756–1832). Nitrogen's role in crop production was not recognized until the mid-19<sup>th</sup> century, initially by Boussingault (1802–1887) and then more thoroughly by von Liebig (1803–1873), who developed the theory of nutrient limitation in plant productivity. Near the end of the 19<sup>th</sup> century, Hellriegel (1831–1895) and Wilfarth (1853–1904) discovered that microbial communities could extract nonreactive N<sub>2</sub> from the atmosphere and convert it into a useable form—biological nitrogen fixation (BNF).

Less than 50 years after N was first identified as an essential nutrient for all plants and animals it was also discovered that the growing demand for food by increasing human populations was exceeding known sources. In 1898, Sir William Crookes, president of the British Association for the Advancement of Science, stated: "All England and all civilized nations stand in deadly peril of not having enough to eat" due to the increasing demands for food and the lack of biologically available nitro-

**Figure 1. Global population trends (36, 53) with key dates for the discovery of N as an element in the periodic table and its role in various biogeochemical processes. Also shown is an estimate of the annual production of Nr by the Haber-Bosch process.**



gen (5). This realization triggered a global search for natural deposits of reactive-N that could be used as fertilizer. At the end of the 19<sup>th</sup> century and the beginning of the 20<sup>th</sup> century, the arid islands of the Pacific and the deserts of South America yielded guano and nitrate deposits which were mined for Nr to grow food in Europe and elsewhere. But other sources were needed. A very timely breakthrough occurred in 1913, invention of a chemical process to convert atmospheric N<sub>2</sub> to NH<sub>3</sub>. The Haber-Bosch process allowed, for the first time, an unlimited supply of nitrogen that could be used to grow food (and make explosives).

After the Haber-Bosch process was invented, the global human population started growing rapidly. While there are many reasons for this, availability of food was an important factor. Today, more than half of the food eaten by the world's peoples is produced using nitrogen fertilizer from the Haber-Bosch process (4).

Energy production also creates Nr, although in contrast to food production, it is produced by accident as a consequence of fossil-fuel combustion. The sources of Nr in energy production are twofold—conversion of atmospheric N<sub>2</sub> to NO<sub>x</sub> and conversion of fossil organic N in the fuel to NO<sub>x</sub>. Both sources result in increased Nr—one from creation of new Nr, and the other from release of sequestered Nr (6). While there are records of coal use in China dating from 500 BC, until the late-19<sup>th</sup> century, most energy was produced from biofuels (e.g. wood). It wasn't until the beginning of the 20<sup>th</sup> century that fossil fuels exceeded biomass fuels in supplying primary energy (7).

## Nr CREATION: PAST AND PRESENT

This section presents the sources and amounts of Nr produced by natural and anthropogenic processes in the past (1890) and the present (1990). 1890 is an appropriate starting point for examination of the N cycle since there was limited Nr creation by human activities. Although the global population was ~ 25% of the current number, the world was primarily agrarian and produced only 2% of the energy and 10% of the grain produced today. Most energy (75%) was provided by biomass fuels; coal provided most of the rest (7). Petroleum and natural-gas production was very limited and was of little consequence relative to the global supply of energy and the creation of Nr as NO<sub>x</sub> through combustion. In total, fossil-fuel combustion created only about 0.6 Tg N yr<sup>-1</sup> in 1890, through production of NO<sub>x</sub> (8).

Crop production was primarily sustained by recycling crop residue and manure on the same land where food was raised. Since the Haber-Bosch process was not yet invented, the only new Nr created by human activities was by legume and rice cultivation (the latter promotes Nr creation because rice cultivation creates an anaerobic environment which enhances nitrogen fixation). While estimates are not available for 1890, Smil estimates that in 1900 cultivation-induced Nr creation was on the order of 15 Tg N yr<sup>-1</sup> (9, and pers. comm.). Additional Nr was mined from guano (~ 0.02 Tg N yr<sup>-1</sup>) and nitrate deposits (~ 0.13 Tg N yr<sup>-1</sup>) (10).

Thus in 1890, the total anthropogenic Nr creation rate was ~ 15 Tg N yr<sup>-1</sup>, almost entirely for food production. In contrast, the natural rate of Nr creation was on the order of 300 Tg N yr<sup>-1</sup>. Terrestrial ecosystems created ~ 100 Tg N yr<sup>-1</sup> (11) and marine ecosystems created ~ 140 Tg N yr<sup>-1</sup> (D. Capone, pers. comm.). An additional ~ 5 Tg N yr<sup>-1</sup> was fixed by lightning (11). On a relative basis for the globe, human activities created about 5% of the total Nr fixed and about 13% when only terrestrial systems are considered.

One century later, the world's population had increased by a factor of ~ 3.5, from about 1.5 to about 5.3 billion, but the global food and energy production increased about 7-fold (10) and 90-fold (7, 12), respectively. Just as was the case in 1890, in 1990 (and now) food production accounts for most of the new Nr created. What changed most since 1890 was the magnitude

of Nr created by humans. Galloway et al. (11) estimated that in 1990 cultivation-induced Nr creation was ~ 40 Tg N yr<sup>-1</sup>. Smil (9) estimated that in the mid-1990s it ranged from 25–41 Tg N yr<sup>-1</sup>, with a mean value of 33 Tg N yr<sup>-1</sup>, which we use in this paper. The Haber-Bosch process, which did not exist in 1890, created ~ 85 Tg N yr<sup>-1</sup> in 1990, mostly for fertilizer (~ 78 Tg N yr<sup>-1</sup>) and the remainder in support of industrial activities such as the manufacture of synthetic fibers, refrigerants, explosives, rocket fuels, nitroparaffins, etc. (9, 12–14).

For energy production, during the period 1890 to 1990, much of the world was transformed from a biofuel to a fossil-fuel economy. The increase in energy production by fossil fuels resulted in increased NO<sub>x</sub> emissions—from ~ 0.6 Tg N yr<sup>-1</sup> in 1890 to ~ 21 Tg N yr<sup>-1</sup> in 1990 (8, 15). By 1990 over 90% of energy production resulted in the creation of new Nr, contrasting to 1890 where very little energy production caused Nr creation.

Thus, in 1990, Nr created by anthropogenic activities was ~ 140 Tg N yr<sup>-1</sup>, a ~ 9-fold increase over 1890, contrasted to a ~ 3.5-fold increase in global population. Coupled with the increase in Nr creation by human activities, was a decrease in natural terrestrial N fixation because of conversion of natural grasslands and forests to croplands, etc. from ~ 100 Tg N yr<sup>-1</sup> to ~ 89 Tg N yr<sup>-1</sup> (C. Cleveland, pers. comm.).

What is the fate of anthropogenic Nr? The immediate fate for the 3 anthropogenic sources is clear—NO<sub>x</sub> from fossil-fuel combustion is emitted directly into the atmosphere; RNH<sub>2</sub> from rice and legume cultivation is incorporated into biomass; NH<sub>3</sub> from the Haber-Bosch process is primarily converted into commercial fertilizer which is applied to agroecosystems to produce food. However, little of this fertilizer-N actually enters the human mouth. To put into context our later discussion of Nr dispersal through environmental reservoirs, it is useful to contrast how much Nr the human body requires, relative to how much Nr is produced by human beings *via* the Haber-Bosch process for fertilizer.

## THE HUMAN BODY VS THE HUMAN BEING: NITROGEN AND FOOD

The human body requires ~ 2 kg N yr<sup>-1</sup> of protein to survive (10). For thousands of years the collective human metabolic N requirement was met by the goods and services provided by unmanaged ecosystems—in essence using the nitrogen produced by naturally occurring BNF. As populations increased and agriculture developed, natural sources of N had to be supplemented with additional sources. Cultivation of rice was the first instance of human-induced BNF through the construction of rice paddies which, when anaerobic, create an environment of enhanced BNF. Archeological evidence suggests that rice has been feeding humans for more than 8000 years (16, 17). Planting of legumes was the second instance of human-induced BNF, with the first substantiated citation of the soybean being in 1000 BC (18).

In contrast to those earliest times of anthropogenic BNF, the 1990 human population of ~ 5.3 billion created about ~ 110 Tg N yr<sup>-1</sup> of Nr for food production, but needed only ~ 11 Tg (at 2 kg person<sup>-1</sup> yr<sup>-1</sup>). What happens to the ~ 100 Tg N yr<sup>-1</sup> that is produced but not needed? Some is consumed as excess Nr, but most is distributed to the environment without entering the human mouth; of course, all the Nr that enters the human mouth is also eventually released to the environment.

There are several steps in the N cycle where leaks to the environment occur: between Haber-Bosch factory and field, between field and crop, between crop and harvest, between harvest and product, and lastly between product and mouth. We illustrate the losses and the reasons by tracking the fate of 100 N atoms formed into fertilizer by the Haber-Bosch process (Fig. 2a). These average estimates are derived from the analyses of Bleken and Bakken (19), Mosier et al. (20), and Smil (4, 12, 21). There is substantial variability in the following numbers but not

in the ultimate result—most of the Nr that is created for food production does not enter the human mouth.

**Factory to field:** Of the 100 N atoms produced, 94 are applied to the agricultural field as fertilizer. The remainder is lost to the environment during fertilizer production, transport (railway, pipeline, truck), storage at the factory, the distributor and the farm (Fig. 2a).

**Field to crop:** Of the 94 atoms applied to the field, only about half are taken up by the crop. The remainder is emitted to the atmosphere (as  $\text{NH}_3$ ,  $\text{NO}$ , or  $\text{N}_2\text{O}$ ), or is lost to groundwater or surface water as primarily nitrate.

**Crop to harvest:** Of the 47 N atoms incorporated into plant protein only 31 are in the harvested crop. The rest are lost from the plant during its growth, or are not harvested (crop residue).

**Harvest to food product:** There are now 31 N atoms remaining. If the harvested material is to be fed to humans, then the production of a food product results in loss of 5 atoms of N due to spoilage, product preparation, etc.

**Product to mouth:** Of the remaining 26 N atoms, 12 are lost due to spoilage and waste. Thus, the efficiency of growing plant protein to feed to humans is about 14%. The remaining 86% of the N is either recycled to agroecosystems or lost to the environment.

There is of course another use of the harvested crop, the production of animal protein. Up to the third step above, the pathway is the same. But we need to revisit the last two steps (Fig. 2b).

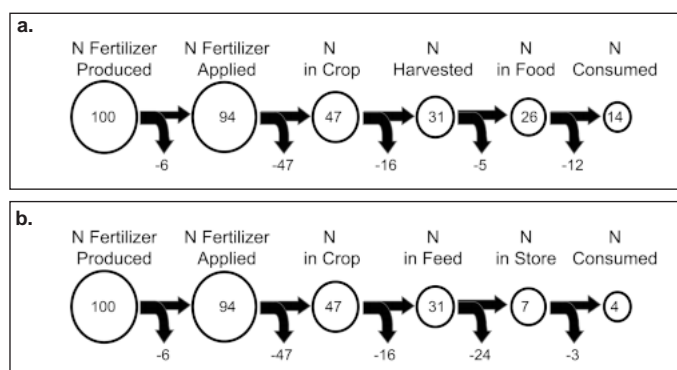
**Harvest to feed product:** There are now 31 N atoms remaining. If the harvested crop is fed to animals which are then prepared into a product, then 24 of the 31 N atoms are lost due to animal metabolism and product preparation.

**Product to mouth:** Of the remaining 7 N atoms that are contained in an animal protein product, 3 are lost due to spoilage and waste. Thus, the efficiency of growing animal protein to feed to humans is about 4%. As above, the remaining 96% of the N is either recycled to agroecosystems (e.g. manure) or lost to the environment.

In summary, the human body needs about 2 kg person<sup>-1</sup> yr<sup>-1</sup>, but human beings (collectively) create ~ 20 kg person<sup>-1</sup> yr<sup>-1</sup> during food production processes. While all of the Nr is distributed to the environment, the Nr that does not enter the human mouth represents an addition of a biogeochemically active element that has both beneficial and detrimental consequences for environmental systems.

## THE GLOBAL N CYCLE: PAST AND PRESENT

This overview of the global N cycle in 1890 (Fig. 3a) and 1990 (Fig. 3b) is based on data from a number of sources. The sources of the data on Nr creation have been discussed previously. Data on atmospheric emissions that are transfers of Nr (as opposed to formation of Nr) are from van Aardenne et al. (8) with the exception of marine  $\text{NH}_3$  emissions (F. J. Dentener, pers. comm.). The atmospheric deposition data are based on Lelieveld and Dentener (22). The estimates of riverine dissolved inorganic nitrogen (DIN) fluxes are from Seitzinger and Kroeze (23). Because of different scales and approaches used to estimate the components of the N cycle in 1890 and 1990, and due to the absence of estimates of denitrification and changes in pool size (e.g. Nr storage), inputs and outputs of Nr are not in balance. In addition, there is substantial uncertainty in some of the estimates (e.g. marine  $\text{NH}_3$  emissions). These characteristics of the analysis notwithstanding, it is clear that the global N cycle underwent a signifi-

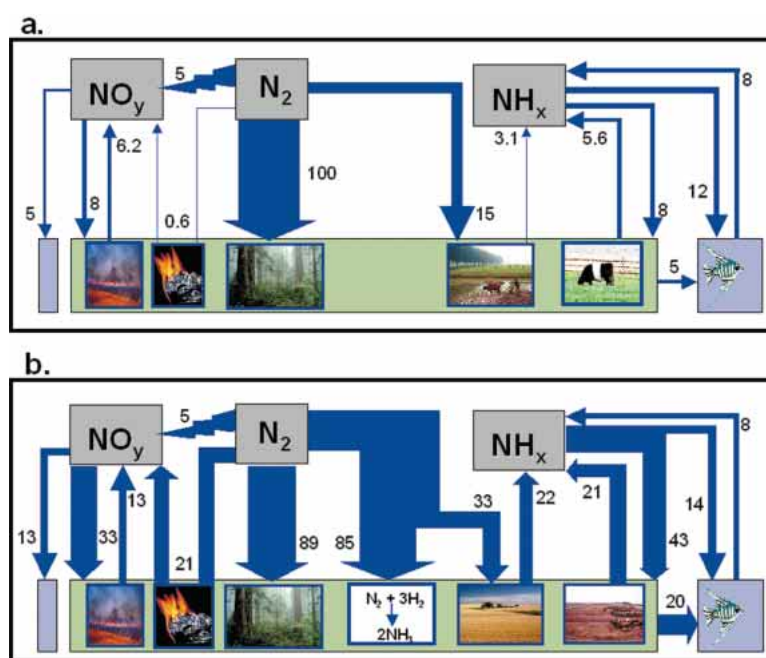


**Figure 2.** The fate of fertilizer N produced by the Haber-Bosch process from the factory to the mouth for (a) vegetarian diet, and (b) carnivorous diet.

cant alteration as a consequence of human action during the 100 years between 1890 and 1990.

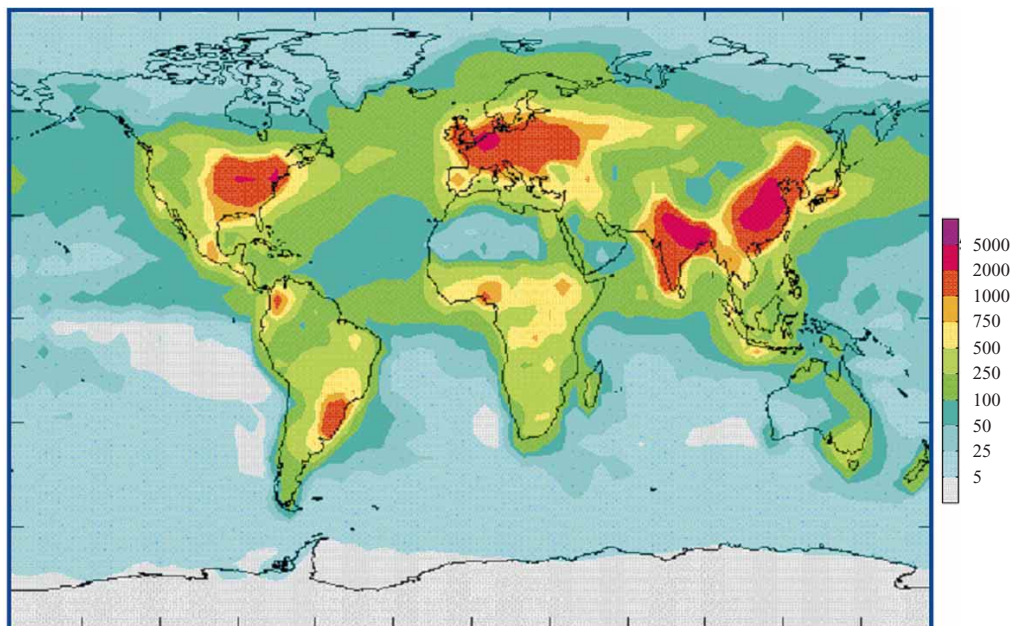
In 1890, creation of Nr was dominated by natural processes (Fig. 3a). The total anthropogenic Nr creation rate was ~ 15 Tg N yr<sup>-1</sup> from legume/rice cultivation (actual year of estimate is 1900) and ~ 0.6 Tg N yr<sup>-1</sup> from fossil-fuel combustion. Natural Nr production in terrestrial systems was ~ 100 Tg N yr<sup>-1</sup>, plus an additional 5 Tg N yr<sup>-1</sup> fixed by lightning. There were limited Nr transfers *via* atmospheric and hydrologic pathways relative to the amount of Nr created by both natural and anthropogenic processes. For terrestrial systems, of the ~ 100 Tg N yr<sup>-1</sup> created, only about ~ 15 Tg N yr<sup>-1</sup> were emitted to the atmosphere as either  $\text{NH}_3$  or  $\text{NO}_x$ . There was limited connection between terrestrial and marine ecosystems. Only about 5 Tg N yr<sup>-1</sup> of DIN were transferred *via* rivers into coastal ecosystems in 1890, and only about 17 Tg N yr<sup>-1</sup> were deposited to the ocean surface.

In 1990, by contrast, creation of Nr was dominated by human activities (Fig. 3b). Natural terrestrial Nr creation was ~ 89 Tg N yr<sup>-1</sup>, compared to cultivation-induced Nr creation of ~ 33 Tg N yr<sup>-1</sup>, Haber-Bosch Nr creation of ~ 85 Tg N yr<sup>-1</sup> and fossil



**Figure 3.** Global nitrogen budget for (a) 1890, and (b) 1990, Tg N yr<sup>-1</sup>. The emissions to the 'NO<sub>y</sub> box' from the 'coal' reflect fossil-fuel combustion. Those from the 'vegetation' include agricultural and natural soil emissions, and combustion of biofuel, biomass (savannah and forests) and agricultural waste. The emissions to the 'NH<sub>x</sub> box' from the 'agricultural field' include emissions from agricultural land and combustion of biofuel, biomass (savannah and forests) and agricultural waste. The NH<sub>x</sub> emissions from the 'cow' and 'feedlot' reflect emissions from animal waste. (For more details, see the section on "Global N Cycle: Past and Present" this paper).

**Figure 4. Global atmospheric deposition of reactive nitrogen (Nr) onto the oceans and continents of the Earth in 1993 ( $\text{mg N m}^{-2} \text{yr}^{-1}$ ). Pers. comm. F.J. Dentener and based upon Lelieveld and Dentener (22).**



fuel combustion which created  $\sim 21 \text{ Tg N yr}^{-1}$ , for a total of  $\sim 140 \text{ Tg N yr}^{-1}$ . Just as there were large changes in Nr creation in 1990 relative to 1890, there were also significant changes in Nr distribution.  $\text{NH}_3$  emissions increased from  $\sim 9 \text{ Tg N yr}^{-1}$  to  $\sim 43 \text{ Tg N yr}^{-1}$  as a consequence of food production;  $\text{NO}_x$  emissions increased from  $\sim 7 \text{ Tg N yr}^{-1}$  to  $\sim 36 \text{ Tg N yr}^{-1}$  from both energy and food production. The increased emissions resulted in widespread distribution of Nr to downwind ecosystems as shown in Figure 4. Transfer of Nr to marine systems also increased. By 1990, riverine DIN fluxes to the coastal ocean had increased to  $20 \text{ Tg N yr}^{-1}$  and atmospheric N deposition to marine regions had increased to  $27 \text{ Tg N yr}^{-1}$ . While evidence suggests that most of the riverine N is denitrified in coastal and shelf environments (24), most of the atmospheric flux is deposited directly to the open ocean, although a portion of the  $27 \text{ Tg N yr}^{-1}$  is deposited to coastal ocean and shelf regions, with significant ecological consequences (25).

A key component missing from Figure 3b is the ultimate fate of the  $\sim 140 \text{ Tg N yr}^{-1}$  Nr created by human action in 1990. On a global basis, Nr created by human action is either accumulated (stored) or is denitrified. Unfortunately, we are not able to estimate the relative importance of these 2 processes, and until we are able, we will not be able to determine the rate at which Nr is accumulating in global systems. We can however, examine case studies of specific landscape types to obtain a very rough estimate of Nr accumulation.

We use 2 case studies here. The first is for a landscape type—global agroecosystems. The second is for specific landscape areas—16 watersheds that drain the northeastern US coast and that range from Maine to Virginia. For each case study, Nr introduced into the system has 3 fates—storage within the system, transfer to another system, or denitrification to  $\text{N}_2$ . Within global agroecosystems, Smil (9) estimates that  $\sim 169 \text{ Tg N yr}^{-1}$  (range of  $151$  to  $186 \text{ Tg N yr}^{-1}$ ) are added to agroecosystems from numerous sources, including fertilizer, recycling of crop residues, and animal manures. Of this total, a mean of  $\sim 14 \text{ Tg N yr}^{-1}$  (range of  $11$  to  $18 \text{ Tg N yr}^{-1}$ ) is denitrified to  $\text{N}_2$ ,  $\sim 8\%$  within agroecosystems. The remainder is recycled back to agroecosystems or is transferred to other systems as food and feed, is discharged to aquatic systems, or is emitted to the atmosphere. There is significant uncertainty about the amount denitrified, and it may in fact be a larger number. For example, van Egmond et al. (26) estimate that for Europe alone, denitrification from agroecosystems is on the order of  $8 \text{ Tg N yr}^{-1}$ , and Howarth et al. (27) estimate that it may be as large as  $\sim 6 \text{ Tg N yr}^{-1}$  for the

United States.

For watershed landscape units composed of a variable mixture of agroecosystems, forests and urban areas, van Breemen et al. (28) found that the average N input from atmospheric deposition, fertilizer use, food/feed import and  $\text{N}_2$  fixation was  $3420 \text{ kgN km}^{-2} \text{yr}^{-1}$ , and of that  $\sim 48\%$  was denitrified ( $\sim 37\%$  in the landscape;  $\sim 11\%$  in rivers). Using a different method of analysis in the same watersheds (for rivers only), Seitzinger et al. (29) estimate that denitrification in rivers may be much larger than that estimated by van Breemen et al. (28). This difference reflects the uncertainty of our knowledge about the rate of conversion of Nr back to  $\text{N}_2$ . It should be noted, that while denitrification is a large sink in the watershed analysis, much of the nitrogen still remained in a reactive form, and of the N that was denitrified, a portion was  $\text{NO}$  and  $\text{N}_2\text{O}$ , both environmentally reactive.

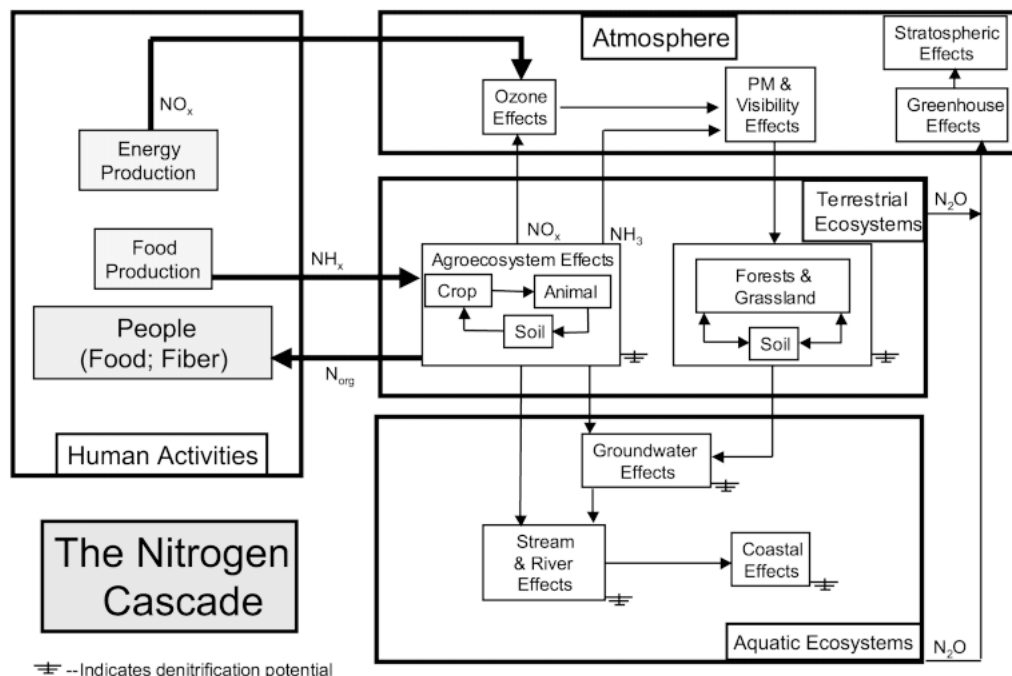
In summary, most anthropogenic Nr is not converted to  $\text{N}_2$  within agroecosystems. In more complex watershed units, a larger portion is converted, especially in aquatic ecosystems. In both examples, the remaining N either accumulates or is dispersed *via* commerce or atmospheric/hydrologic transport to other environmental systems where it may be stored, denitrified or further dispersed.

## THE NITROGEN CASCADE

About 1965, the rate of human creation of new Nr began to exceed natural terrestrial creation of Nr. Since conversion of Nr back to  $\text{N}_2$  by denitrification appears not to be keeping pace with creation of new Nr, it is accumulating in various environmental reservoirs (e.g. atmosphere, soils, waters). The accumulation of Nr in environmental reservoirs has significant effects on humans (30) and ecosystems (25, 26, 31).

There are large uncertainties regarding the rates of Nr accumulation in various reservoirs. This limits our ability to determine the temporal and spatial distribution of environmental effects. These uncertainties are even more significant because of the sequential nature of the effects of Nr on environmental processes. This sequence of transfers, transformations, and environmental effects is referred to as the nitrogen cascade (32, and Galloway et al., unpublished). As depicted in Figure 5, there are 2 primary sources of Nr— $\text{NH}_x$  creation mainly from food production, and  $\text{NO}_x$  emissions mainly from energy production. A single atom of new  $\text{NH}_x$  or  $\text{NO}_x$  can alter a wide array of biogeochemical processes and exchanges among environmental reservoirs. These processes include:

Figure 5. The nitrogen cascade illustrates the movement of human-produced reactive nitrogen (Nr) as it cycles through various environmental reservoirs in the atmosphere, terrestrial ecosystems, and aquatic ecosystems of the Earth (Galloway et al. unpubl.).



- Photochemical transformations and greenhouse effects within the atmosphere.
- Biological transformations and effects within terrestrial ecosystems.
- Biological transformations and effects within freshwater and coastal marine environments.

This sequence of Nr effects is somewhat analogous to the ability of NO to keep regenerating in the stratosphere and sequentially destroy one ozone molecule after another.

As new Nr cascades through various environmental reservoirs, it contributes to a wide variety of changes that impact humans and ecosystems in different ways in various parts of the world. Some of these changes are beneficial for society; others are less so or are detrimental. Table 1 provides a detailed list of these beneficial and detrimental effects as described in the *Summary Statement for the Second International Nitrogen Conference* (33). In addition to their great variety, another intriguing aspect of these effects is their linkage through the biogeochemical processes of the nitrogen cascade. For example, as shown in Figure 5, before being converted back to nonreactive N<sub>2</sub>, a given Nr atom can in sequence:

- Increase ozone concentrations in the troposphere.
- Decrease atmospheric visibility and increase concentrations of PM<sub>2.5</sub> particles.
- Increase precipitation acidity.
- Increase soil acidity.
- Increase or decrease forest productivity.
- Increase surface water acidity.
- Increase hypoxia in coastal waters.
- Increase greenhouse warming.
- Decrease stratospheric ozone.

A principle feature of the cascade is the accumulation rate of Nr in environmental systems. This is one of the most important research questions associated with the impact of humans on the nitrogen cycle.

## REGIONAL ANALYSIS

To assess regional variability of Nr creation and distribution we divide the world into several regions, Africa, Asia, Europe, Latin America, North America, and Oceania, as defined by United Nations Food and Agriculture Organization (FAO) (14). For the mid-1990s in each region, we have determined the amount of Nr created by fertilizer production (14), fossil-fuel combustion (F. J. Dentener, pers. comm.) and rice/legume cultivation (14).

Most fertilizer production occurred in Asia (40.1 Tg N yr<sup>-1</sup>), Europe (21.6 Tg N yr<sup>-1</sup>) and North America (18.3 Tg N yr<sup>-1</sup>) with smaller amounts in Latin America, Africa, and Oceania (3.2, 2.5 and 0.4 Tg N yr<sup>-1</sup>, respectively). The other Nr creation process involved in food production is cultivation-induced BNF. Smil (9) estimates that ~ 33 Tg N yr<sup>-1</sup> were produced by cultivation globally in the mid-1990s. The FAO has country and regional breakdowns of harvested areas for seed legumes, rice and sugar cane that we use with BNF rates (9) that we use to estimate regional BNF. We calculate these BNF rates from cultivation of

**Table 1. Beneficial and detrimental effects of reactive nitrogen (Nr).**

<p>Direct effects of Nr on human health include:</p> <ul style="list-style-type: none"> <li>Increased yields and nutritional quality of the foods needed to meet dietary requirements and food preferences for growing populations.</li> <li>Respiratory and cardiac disease induced by exposure to high concentrations of ozone and fine particulate matter.</li> <li>Nitrate and nitrite contamination of drinking water leading to the "blue-baby syndrome" and certain types of cancer.</li> <li>Blooms of toxic algae, with resultant injury to humans.</li> </ul>
<p>Direct effects of Nr on ecosystems include:</p> <ul style="list-style-type: none"> <li>Increased productivity of Nr-limited natural ecosystems.</li> <li>Ozone-induced injury to crop, forest, and natural ecosystems and predisposition to attack by pathogens and insects.</li> <li>Acidification and eutrophication effects on forests, soils, and freshwater aquatic ecosystems.</li> <li>Eutrophication and hypoxia in coastal ecosystems.</li> <li>N saturation of soils in forests and other natural ecosystems.</li> <li>Biodiversity losses in terrestrial and aquatic ecosystems and invasions by N-loving weeds.</li> <li>Changes in abundance of beneficial soil organisms that alter ecosystem functions.</li> </ul>
<p>Indirect effects of Nr on other societal values include:</p> <ul style="list-style-type: none"> <li>Increased wealth and well being of human populations in many parts of the world.</li> <li>Significant changes in patterns of land use.</li> <li>Regional hazes that decrease visibility at scenic vistas and airports.</li> <li>Depletion of stratospheric ozone by N<sub>2</sub>O emissions.</li> <li>Global climate change induced by emissions of N<sub>2</sub>O and formation of tropospheric ozone.</li> <li>Damage to useful materials and cultural artifacts by ozone, other oxidants, and acid deposition.</li> <li>Long-distance transport of Nr which causes harmful effects in countries distant from emission sources and/or increased background concentrations of ozone and fine particulate matter.</li> </ul>
<p>In addition to these effects, it is important to recognize that:</p> <ul style="list-style-type: none"> <li>The magnitude of Nr flux often determines whether effects are beneficial or detrimental.</li> <li>All of these effects are linked by biogeochemical circulation pathways of Nr.</li> <li>Nr is easily transformed among reduced and oxidized forms in many systems.</li> <li>Nr is easily distributed by hydrologic and atmospheric transport processes.</li> </ul>

**Table 2. Nr creation in mid-1990s for various regions of the world, Tg N yr<sup>-1</sup>.**

World Regions	Fertilizer production	Cultivation	Combustion	Net import/export	Total
Africa	2.5	1.8	0.8	0.2	5.3
Asia	40.1	13.7	6.4	8.7	68.9
Europe + FSU	21.6	3.9	6.6	-5.6	26.5
Latin America	3.2	5.0	1.4	-0.2	9.4
North America	18.3	6.0	7.4	-3.3	28.4
Oceania	0.4	1.1	0.4	0.3	2.2
World	~ 86	~ 30	~ 23	0.1	~ 140

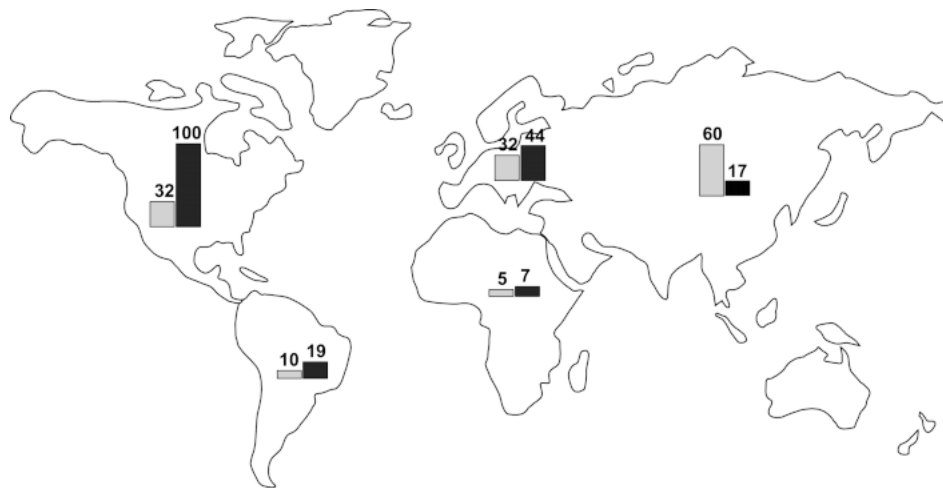
**Table 3. Regional per capita Nr creation in mid-1990s during food and energy production, kg N person<sup>-1</sup> yr<sup>-1</sup>.**

Regions	Food	Energy	Total
Africa	5.7	1.1	6.8
Asia	15	1.8	17
Europe & FSU	35	9.1	44
Latin America	16	2.8	19
North America	80	24	100
Oceania	50	13	63
World	20	3.9	24

seed legumes, rice, and sugar cane: Africa, 1.8; Asia, 9.2; Europe + Former Soviet Union (FSU), 0.4; Latin America, 3.0; North America, 2.5; Oceania 0.3 Tg N yr<sup>-1</sup>). However, FAO does not have equivalent data for N fixation by leguminous cover crops and non-Rhizobium diazotrophs that can be used to divide by region the ~ 14 Tg N yr<sup>-1</sup> produced annually from these 2 sources (9). But, based on an analysis of fixation for selected major countries, Smil (V. Smil, pers. comm.) suggests that the apportionment of the 14 Tg N yr<sup>-1</sup> among the regions is ~ 4.5, 3.5, 2.0, 3.5, and 0.8 for Asia, Europe + FSU, Latin America, North America, and Oceania, respectively, with an uncertainty of about 25% around these estimates. Summing, we found that cultivation-induced BNF was focused in Asia (~ 13.7 Tg N yr<sup>-1</sup>), with more modest creation rates in North America, Europe + FSU, and Latin America (~ 6.0, ~ 3.9 and ~ 5.0 Tg N yr<sup>-1</sup>, respectively). Africa and Oceania created ~ < 2 Tg N yr<sup>-1</sup>. Nr creation by fossil fuel combustion occurred primarily in North America, Europe and Asia (~ 7.4, ~ 6.6 and ~ 6.4 Tg N yr<sup>-1</sup>, respectively). Latin America, Africa and Oceania by comparison had more modest Nr creation rates, with each being < 1.5 Tg N yr<sup>-1</sup> (Table 2).

There is an additional Nr source for some regions. A portion of the Nr produced within a region can be exported to other regions. Exports of N-containing fertilizer, plant material (e.g. grain) and meat from one region will be a source for another region. To determine the net gain (loss) of N commodities for the regions we compiled import and export data from the FAO database (14). Fertilizer was the commodity most often exchanged between regions. In 1995, the global production of N fertilizers was ~ 86 Tg N yr<sup>-1</sup>. Of this amount, ~ 24.9 Tg N yr<sup>-1</sup> were exported to other regions. Over half of the exports were from Europe (~ 13.2 Tg N yr<sup>-1</sup>). Other regions with significant exports were Asia (~ 10.7 Tg N yr<sup>-1</sup>) and North America (~ 5.2 Tg N yr<sup>-1</sup>). The primary receiving regions were Asia (~ 7.6 Tg N yr<sup>-1</sup>) and Europe (~ 6.6 Tg N yr<sup>-1</sup>). Thus, while about ~ 30% of the fertilizer N produced was exported, the only region that had a large net loss was Europe (~ 6.6 Tg N yr<sup>-1</sup>). The largest net gain of any region was for Asia (~ 6.4 Tg N yr<sup>-1</sup>) (Table 2).

The next most frequently exchanged commodity was plant material, mostly cereal grains. Asia, Europe and Africa had net gains in N-containing plant material (~ 2.2, ~ 1.0, ~ 0.5 Tg N yr<sup>-1</sup>, respectively), while North America, Latin America, and Oceania had net losses (~ 2.8, ~ 0.8, ~ 0.2 Tg N yr<sup>-1</sup>, respectively). Net meat exchange was < 0.1 Tg N yr<sup>-1</sup> for each region. Summing all three categories (Table 2), Asia gained the most Nr—~ 8.7 Tg N yr<sup>-1</sup>. While Oceania and Africa also gained N, the gains were small (~ 0.3 Tg N yr<sup>-1</sup> and ~ 0.2 Tg N yr<sup>-1</sup>, respectively). Europe and North America had net losses of ~ 5.6 and ~ 3.3 Tg N yr<sup>-1</sup>, respectively. Latin America had a small net loss. When these net import/exports are added to the Nr creation in each region (Table 2), North America, with ~ 5% of the

**Figure 6. Comparison of reactive nitrogen (Nr) creation rates for various regions of the world. The left bar shows the total amount of Nr created in each region in 1995 (Tg N yr<sup>-1</sup>). The right bar shows the amount of Nr created in each region in 1995 on a per capita basis (kg N person<sup>-1</sup> yr<sup>-1</sup>).**

world's population, is responsible for creation of ~ 20% of the world's Nr. Africa, with ~ 13% of the world's population, is responsible for ~ 6% of the world's Nr.

Given these regional differences, it is also interesting to express Nr creation and use on a per capita basis (calculated from Table 2 (not including net import/export) and FAO population data (14)), to illustrate the average amount of Nr mobilized per person, by region (Table 3). At one extreme North America mobilizes ~ 100 kg N person<sup>-1</sup> yr<sup>-1</sup>; at the other extreme, Africa, mobilizes about an order of magnitude less, ~ 7 kgN person<sup>-1</sup> yr<sup>-1</sup> (Fig. 6). In all regions, food production is larger than energy production, and the primary cause for the regional differences are the amounts of N mobilized per capita in the production of food. In North America, it is ~ 80 kgN person<sup>-1</sup> yr<sup>-1</sup>; in Africa it is ~ 6 kgN person<sup>-1</sup> yr<sup>-1</sup>. Remembering back to the section on 'Body vs. Being', these values show which regions are responsible for Nr creation in excess of what is needed for human body sustenance.

This regional analysis focuses on Nr creation and interregional exchange through commerce. Very detailed regional case studies are presented for Europe (26), United States (27) and Asia (34) in this issue of *Ambio*.

## FUTURE

Nr creation by human action will continue to increase in the future as global populations grow. Even after they peak, perhaps sometime in this century (35), Nr creation will still most likely continue to increase due to growth in per capita resource use. How high will the Nr-creation rate go? In 1990, it was ~ 140 Tg N yr<sup>-1</sup>, and the average per capita Nr-creation rate was ~ 24 kg N person<sup>-1</sup> yr<sup>-1</sup>, and ranged from ~ 7 kg N person<sup>-1</sup> yr<sup>-1</sup> in Africa to ~ 100 kg N person<sup>-1</sup> yr<sup>-1</sup> in North America (Fig. 6).

If the global population peaks at ~ 8.9 billion people (estimate of global population in 2050; United Nations (36)) and if all people had the same per capita Nr-creation rate from food and energy production as North America in 1990 (~ 100 kg N person<sup>-1</sup> yr<sup>-1</sup>), then the total Nr-creation rate would be ~ 900 Tg N yr<sup>-1</sup>,

with about half occurring in Asia (Fig. 7). Given the concerns about Nr, it is very unlikely that this value will be reached. What the final maximum Nr-creation rate turns out to be, however, will depend to a very large extent on how the world manages its use of nitrogen for food production and its control of N in energy production.

Thus, there is ample opportunity for creative management of Nr in the development of strategies that will allow for optimization of food and energy production while also optimizing environmental health. Several papers in this volume address the issue of Nr management to maximize food and energy production, and protect the health of people and ecosystems. Management of energy producing systems is discussed by Moomaw (37) and Bradley and Jones (38). For food production, management issues are discussed in Cassman et al. (39), Fixen and West (40), Oenema and Pietrzak (41), Roy et al. (42) and Smil (21). Policy options are presented in some of these papers as well as in Melillo and Cowling (43).

As is noted in other papers, for management to be effective, it must take into account all aspects of Nr. The next section presents the case for developing a Total Nr Approach for management of Nr.

#### DEVELOPMENT OF A CONCEPT OF OPTIMUM Nr MANAGEMENT FOR SOCIETY

In his most famous book, "Future Shock," Toffler (44) identified 3 different types of futures, that he believed innovative democratic societies should consider very carefully:

- i) *Probable futures*: Hopes and aspirations of society that are largely an extension of a "business as usual" sense of what the future might hold.
- ii) *Possible futures*: Exploration of all possible outcomes that a given society might wish to explore as possibilities for its future.
- iii) *Preferable futures*: Optimum outcomes that probably can be achieved only as a result of focused and well-disciplined efforts to fulfill mutually agreed upon goals and dreams which are consonant with the natural and human resources available to society.

An excellent example of efforts to evaluate 'preferable futures' is provided by the computer simulation game, NitroGenius (45), developed for the Dutch government, and presented at the Second International Nitrogen Conference.

In evaluating alternative choices about management of air and water quality in the context of other important societal goals, enlightened societies will want to consider Toffler's suggestions and thus go beyond "business as usual" perspectives, look earnestly at a wide range of "possibilities," and work hard to define and implement "preferable" options that are both prudent and realistic for the long-term as well as for the short-term futures of society (46, 47).

So far, most of the voluntary recommended management practices and the mandated rules and regulations for management of Nr have been developed and administered separately. For example, air emissions of NO<sub>x</sub> were first regulated because NO<sub>x</sub> is an important precursor of ozone and later because it also contributes to acidification of soils and surface waters. Similarly, air emissions of ammonia first became a pollutant of concern because ammonia contributes to acidification processes. Also, most guidance for prevention of nitrogen emissions from confined animal feeding operations in North America have been developed and administered with emphasis on direct discharges to



**Figure 7. Comparison of contemporary and possible future reactive nitrogen (Nr) creation rates in various regions of the world. The left bar shows the total amount of Nr created in each region in 1995 (Tg N yr<sup>-1</sup>). The right bar shows the amount of Nr that would be created in each region at the time of maximum projected human population of the Earth (~ 8.9 billion people) assuming that the average Nr creation rate in North America in 1995 (~ 100 kg N person<sup>-1</sup> yr<sup>-1</sup>) was achieved in all regions of the world.**

watercourses with little or no concern about the associated (and much larger) air emissions of volatile ammonia and amines. All oxidized, reduced, and organic forms of Nr participate in a variety of chemical and physical transformations in the atmosphere. As indicated in Table 1, they also can have a long series of beneficial and detrimental biological effects once they are transferred into terrestrial and aquatic ecosystems as indicated in the nitrogen cascade (Fig. 5).

The time has come to develop and implement a Total Reactive Nitrogen Approach (Total Nr Approach), rather than continue to consider nitrogen-oxide pollution and ammonia pollution in isolation from each other and from other aspects of air- and water-quality management. A Total Nr Approach is firmly grounded in the following biological principles (48–50):

- i) Although there are transitory differences in rates of uptake and assimilation, all oxidized, reduced, and organic forms of Nr have substantially similar influences on the general productivity of the terrestrial, aquatic, and livestock-dominated ecosystems systems in which they are deposited and assimilated. This is true because at least one or another (and sometimes many) of the various plants, animals, microbes, and insects in terrestrial and aquatic ecosystems take up all oxidized, reduced, and organic forms of Nr. After initial uptake and assimilation, these various forms of Nr are readily transformed and exchanged with other organisms and compartments within a given landscape or watershed.
- ii) When transferred into ecosystems in less than optimal amounts, oxidized, reduced, organic forms of Nr are biologically available and increase the productivity of the system.
- iii) When applied in more than optimal amounts, however, all biologically active forms of Nr contribute to the wide variety of detrimental effects listed in Table 1.
- iv) The total supply of Nr in terrestrial and aquatic ecosystems is a complex function of the atmospheric, terrestrial, and aquatic transfer, and transformation processes outlined in the nitrogen cascade (Fig. 5).

A Total Nr Approach is especially important in the context of recent international negotiations: i) The recommendations of Grennfelt et al. (51) for multiple-pollutant perspectives in the new nitrogen protocol; and ii) acceptance of the "multiple-pollutant/multiple-effects" (Gothenburg Protocol) by the now 31 North American and European nations that signed this protocol following its final negotiation in December 1999 (52).

#### CONCLUSIONS

Over the course of about 2 centuries, human involvement with N progressed from its discovery in the late-18<sup>th</sup> century to hu-

man dominance of its terrestrial cycle in the late-20<sup>th</sup> century. The production of food and energy, so critical to human survival, has resulted in substantial increases in the amount of Nr in environmental reservoirs. These increases have a number of effects, both positive and negative, which due to the biogeochemical characteristics of Nr, can occur in sequence as Nr cascades through the environment.

While the benefits of increasing amounts of Nr are numerous, all of the Nr that is created by human action is eventually released into the environment. A critical topic for research is de-

termination of the fate of this Nr. Where is it going? How much is stored? How much is denitrified? We will not be able to determine the impact of Nr mobilization on human and environmental systems until we can account for its fate. Another critical topic for research is how can we manage Nr more effectively? Specifically, how can we eliminate the creation of Nr by energy production? How can we decrease the amount of nitrogen released during food production?

These critical issues will require a new way of examining how people manage the environment and their use of resources.

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