Transformation of the Nitrogen Cycle: Recent Trends, Questions, and Potential Solutions

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Humans continue to transform the global nitrogen cycle at a record pace, reflecting an increased combustion of fossil fuels, growing demand for nitrogen in agriculture and industry, and pervasive inefficiencies in its use. Much anthropogenic nitrogen is lost to air, water, and land to cause a cascade of environmental and human health problems. Simultaneously, food production in some parts of the world is nitrogen-deficient, highlighting inequities in the distribution of nitrogen-containing fertilizers. Optimizing the need for a key human resource while minimizing its negative consequences requires an integrated interdisciplinary approach and the development of strategies to decrease nitrogen-containing waste.

Our understanding of reactive nitrogen (Nr) (1) and the N cycle has shifted from how to promote food production to a realization that agricultural intensification damages environmental systems (2). Since 1970, world population has increased by 78% and reactive nitrogen creation has increased by 120%. In 1970, Delwiche stated, "The ingenuity that has been used to feed a growing world population will have to be matched quickly by an effort to keep the nitrogen cycle in reasonable balance" (3). Thirty-five years later, Dobermann and Cassman pointed out, "Failure to arrest the decrease in cereal crop area and to improve nitrogen use efficiency in the world's most important agricultural systems will likely cause severe damage to environmental services at local, regional, and global scales due to a large increase in reactive N load in the environment (4)."

It is clear that an optimum has not been achieved. In some parts of the world, Nr has been used to create an excess of food and a growing prevalence of unhealthy diets, while also con-

tributing to a host of environmental problems (2, 5–7). Yet, other world regions lack sufficient Nr to meet even the most basic caloric demands of hundreds of millions of people (8).

Major research and management challenges remain and are becoming ever more pressing as the creation and use of Nr continues to accelerate. Although diverse management strategies are necessary, they are also possible, and we believe a more favorable balance between the benefits and unwanted consequences of Nr can be achieved.

Continued Acceleration of Nr Creation

Nr creation continues to increase every year. It is dominated by agricultural activities, but fossil fuel energy plays an important role, and the growing prevalence of biofuels is adding a new and rapidly changing dimension. From 1860 to 1995, energy and food production increased steadily on both an absolute and per capita basis; Nr creation also increased from ~15 Tg N in 1860 to 156 Tg N in 1995. The change was enormous, and it increased further from 156 Tg N yr^{-1} in 1995 to 187 Tg N yr^{-1} in 2005, in large part because cereal production increased from 1897 to 2270 million tons (20%), and meat production increased from 207 to 260 million tons (26%) (9). These rising agricultural demands were sustained by a rise in Nr creation by the Haber-Bosch process from 100 Tg N yr^{-1} to 121 Tg N yr^{-1} (20%) (9). Cultivation-induced biological nitrogen fixation (C-BNF) occurs in several agricultural systems, with crop, pasture, and fodder legumes being the most important (10). The C-BNF estimate for 1995 was 31.5 Tg N (5) and, because of the increase in soybean and meat production over the past decade, we estimate that in 2005 C-BNF was 40 Tg N. There is substantial uncertainty in this value, and this is a critical area where more precise data are needed. In parallel, primary

commercial energy production by coal, natural gas, and petroleum combustion increased from 8543 million tons of oil equivalents (mtoe) to 10,600 mtoe (24%), much of it in the developing world (11). However, decreases in NOx emissions in the developed world, among other reasons, led to a relatively constant global creation rate of Nr–NOx of ~25 Tg N yr^{-1} from 1995 to 2000 (12), and we assume for the purpose of discussion that this value also holds for 2005.

Finally, an important but poorly understood aspect of N mobilization is industrial Nr use. NH$_3$ from the Haber-Bosch process is used as a raw material to create multiple products, including nylon, plastics, resins, glues, melamine, animal/fish/shrimp feed supplements, and explosives. In 2005, ~23 Tg N was used for chemical production (13), accounting for 20% of Haber-Bosch Nr, but little is known about the fate of Nr used in these industrial activities.

Nr Distribution Patterns Are Changing

In 2004, ~45 Tg N of the ~187 Tg N of Nr created was traded internationally (Fig. 1), and in the preceding decade, global trade of N commodities increased twice as fast as the rate of Nr creation. Unlike aquatic or atmospheric transport, where Nr is diluted to varying degrees, commerce typically results in injection of Nr to ecosystems in more concentrated doses. Although this has the potential to cause greater damage to a smaller region, it also allows the possibility of greater control over Nr release. However, the rise in international trade is posing new socioeconomic questions, such as who pays for environmental damage associated with Nr losses (14). Regions that consume N-containing products, such as meat and milk, may be far removed from regions that produce the commodity and thus do not have to bear the environmental cost of the production. For example, in 1910, The Netherlands used 13 k tonne of fertilizer N yr^{-1} to produce food for its population of 6 million. In 1999, for the same agricultural area, 400 k tonne N yr^{-1} fertilizer N was used, and the yields were enough to feed 32 million people, only half of whom lived in The Netherlands. The rest of the food, and the Nr it contained, was exported, whereas the N lost in the food-production process remained in the Dutch environment, causing increased groundwater pollution, ambient ammonia and particle emissions, and nitrogen deposition (15). Similarly, areas of Latin America are bearing the cost of land conversion for soy that is fueling rising meat consumption in Asia (14).

On a global basis, atmospheric transport and subsequent deposition has become the dominant Nr distribution process. It is estimated that in 1860, 34 Tg N yr^{-1} of Nr was emitted as NOx and NH$_3$ and then deposited to the Earth's surface as NOy and NHx; in 1995, it had increased to 100 Tg N yr^{-1}; by 2050, it is projected to be 200 Tg N yr^{-1} (5). N deposition to ecosystems in the absence of human influence is generally

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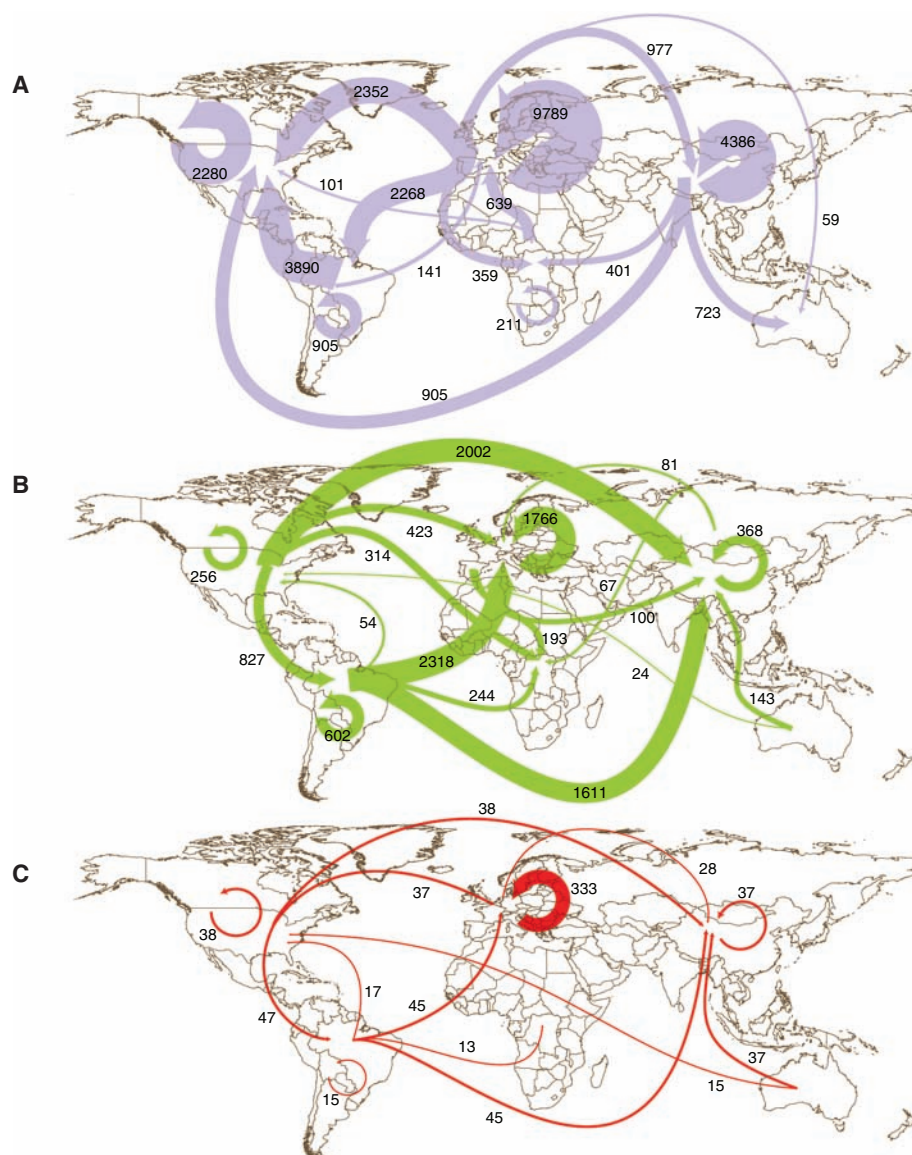
Fig. 1. N contained in internationally traded (**A**) fertilizer (31 Tg N), (**B**) grain (12 Tg N), and (**C**) meat (0.8 Tg N). Data are for 2004 and are in units of thousand of tons. Minimum requirements for drawing a line are 50,000 tons N, 20,000 tons N, and 10,000 tons N for fertilizer, grain, and meat, respectively (*42*).

mental responses (*20*). Yet, we also know that Nr creation is essential to support a burgeoning human population (*21*) and that hundreds of millions of people still suffer from a "fertilizer deficit" (*8*). Finally, we know that environmental changes wrought by excess Nr can feed back to affect human health and welfare, both directly, for example through increased production of atmospheric particulate matter, and indirectly through impacts on food production (*6*). Thus, the grandest overall challenge posed by a changing N cycle is how to maximize the benefits of anthropogenic Nr while minimizing its unwanted consequences (see www.initrogen.org).

Although the role of Nr in multiple aspects of environmental change is undeniable, important research questions remain unresolved. We have identified five broad categories of questions that are priorities for future research.

What is the ultimate fate of Nr? Although data on the creation of anthropogenic Nr are relatively well constrained, those on its fate are uncertain. For example, in the mid-1990s, the fate of only 35% of Nr inputs to the terrestrial biosphere was relatively well known: 18% was exported to and denitrified in coastal ecosystems, 13% was deposited to the ocean via the marine atmosphere, and 4% was emitted as N_2O (*5*). Thus, the majority (65%) either accumulated in soils, vegetation, and groundwater or was denitrified to dinitrogen (N_2), but the uncertainty of those estimates remains large at every scale.

Even with these uncertainties, it is likely that denitrification is an important Nr sink. The first spatially explicit pattern of denitrification from soils to the coastal ocean suggested that more than 80% of denitrification is occurring in soils and freshwater systems (groundwater, rivers, lakes, and reservoirs). The bulk of the remainder (~15%) appears to occur in continental shelf sediments, thus indicating that rivers, although important sources to coastal systems, are typically small sources of Nr to the open ocean, even in heavily altered regions (*22*).

There is a growing database on Nr riverine fluxes, and several models are available that relate watershed characteristics to Nr flux (*23, 24*). Even with these advances, some of the largest uncertainties in measuring denitrification rates are in upland terrestrial systems, which seem to account for a considerable, but unknown, Nr "sink." Nr inputs to these systems continue to rise, however, so the question is whether the fraction of N exported to the coasts will remain small or whether upland "sinks" will saturate to allow greater N-fueled coastal change.

Rising levels of atmospheric deposition also lend urgency to multiple questions about the fate of Nr. Ultimately, the fate of Nr that enters terrestrial systems appears to be under strong climatic control (*25*), an interaction that helps explain regional differences in N export and that should be considered in forecasts of future N-cycle dynamics. The fate and impacts of Nr are also often dependent on its chemical form,

~0.5 kg N ha^{-1} yr^{-1} or less (*16*). There are now large regions of the world where average N deposition rates exceed 10 kg N ha^{-1} yr^{-1}, greater than an order of magnitude increase compared with natural rates. By 2050, this may double, with some regions reaching 50 kg N ha^{-1} yr^{-1} (*5*), and landscape-level inputs may be much larger, especially for forest ecosystems (*17*). These rates are well in excess of the critical loads that have detrimental impacts on receiving ecosystems (*18*) (Fig. 2). Critical loads for the open ocean have not been calculated, but Duce *et al.* (*19*) conclude that the increasing amounts of atmospheric anthropogenic Nr entering the ocean could increase annual new marine biological production by ~3% and increase the emission of N_2O to the atmosphere by ~1.6 Tg N yr^{-1}.

Given the growing importance of the atmosphere in Nr distribution, it is critical to get a better understanding of emissions rates. There is a relatively good understanding of NOx emissions from fossil fuel combustion, but less so from biomass burning and soil emissions. The largest uncertainties are in the NH_3 emissions rates, from all sources, on all scales (*17*). There are also critical questions about the fate and impact of the N deposited to terrestrial, freshwater, and marine realms.

Vexing Questions

Nr creation is still accelerating, a trend unlikely to change in the near future. The additional anthropogenic Nr affects climate, the chemistry of the atmosphere, and the composition and function of terrestrial and aquatic ecosystems (*2*). Moreover, because a single molecule of reactive N can "cascade" through the environment, it can contribute to more than one of these environ-

further highlighting the need to better resolve changing inputs of oxidized versus reduced forms of Nr.

What are the net climate effects of increasing Nr? Nitrogen is both influenced by and affects climate; the net contributions of anthropogenic Nr to a changing climate remain widely debated (*17, 26*). Nr can directly increase radiative forcing in the troposphere, principally through the production of N_2O and tropospheric O_3, but atmospheric Nr can also have cooling effects (*26*), largely through tropospheric aerosols and stratospheric O_3 declines. Moreover, Nr has strong interactions with the carbon (C) cycle that can have global-scale effects on atmospheric carbon dioxide (CO_2) and methane (CH_4) (*27, 28*).

Elevated N deposition may stimulate plant growth in N-limited regions and cause substantial CO_2 uptake in Northern Hemisphere forests, although the size is controversial (see SOM text). N-driven C storage in nonforested or agricultural systems appears modest at best, and N-fueled

ics are also home to the bulk of the planet's terrestrial and freshwater biodiversity. Given that elevated Nr inputs are known to drive biodiversity losses in higher latitude ecosystems (*32*), the projected trends in tropical regions are cause for concern (*33*).

Most biological N fixation in terrestrial systems occurs in tropical regions; this, combined with the widespread existence of phosphorus- and cation-deficient soils causes many tropical ecosystems to exist in a relatively N-rich state (*34*). Thus, at least in theory, the response of these systems to additional N inputs could be very different from those of temperate ecosystems and could result in rapid N losses to air and water, soil cation depletion, and reduced C uptake (*34*). Yet, data on such ecosystem responses and their translation into effects on community structure and biodiversity loss remain notably rare.

How does Nr affect human health? The ability to fix N on large scales is unquestionably a boon to humanity. Perhaps 40% of the
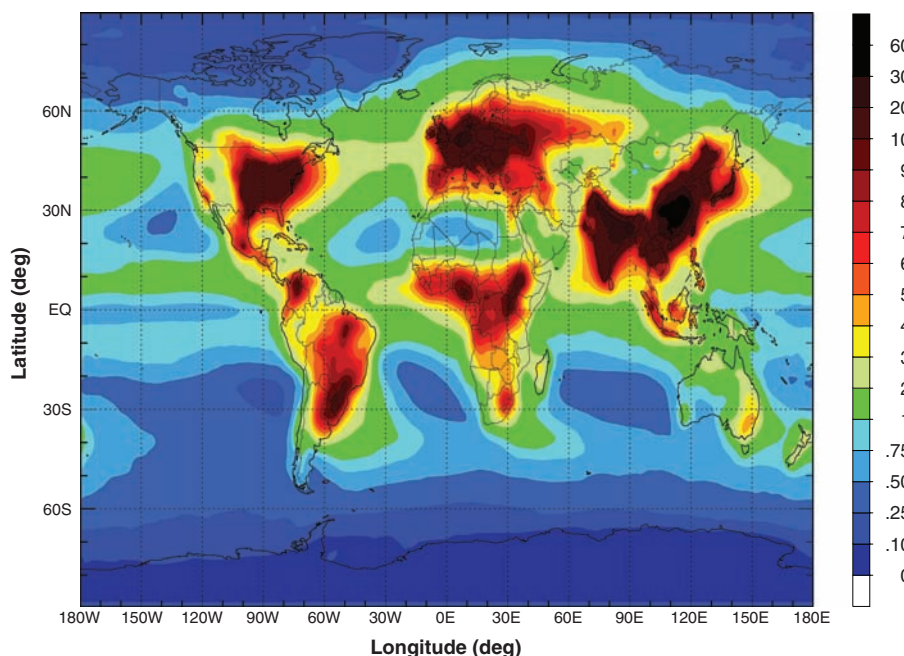
and cause substantial losses in agricultural productivity (*35*). Nitric oxide and ammonia emissions fuel fine-particle and tropospheric O_3 formation, which exacerbate pulmonary disease (*6*). The health consequences of drinking water with elevated nitrate levels, including cancer and reproductive risks, remain poorly known but are important to resolve (*36*). Excess N in the environment may also change the prevalence of important infectious diseases, including malaria, West Nile virus, cholera, and schistosomiasis (*37*). Yet, in some regions with heavy infectious disease burdens, Nr is needed for adequate nutrition to mount effective immune responses to infection. As countries industrialized during the past century, improved nutrition alone reduced the threat from infectious diseases (*6*).

How will biofuel development alter the N cycle? The rapid development of biofuels has created an entirely new link between human activities and the global N cycle, but the full suite of connections is not well resolved. Currently, much of the world's biofuels are produced from corn in the United States or sugar cane in Brazil. U.S. corn covers nearly 29 million ha and is fertilized by an average of 160 kg N ha^{-1} yr^{-1}; Brazilian cane covers ~ 7 million ha and receives an average of 100 kg N ha^{-1} yr^{-1}. As with many intensive agricultural systems, N fertilizer use efficiency in Brazilian sugar cane is low: Only ~ 30% ends up in plant tissues (*38*). Thus, most of the applied N reaches the environment, and because the sugar cane area is predicted to double in Brazil by 2016, the biofuel industry will contribute to a rapidly changing tropical N cycle (*38*). Consequently, N-intensive biofuels could cancel out any CO_2 savings by contributing to both N_2O and tropospheric O_3 production (*39*). Second-generation biofuels will use more woody biomass from year-round crops and production forests and tend to have much higher conversion efficiencies. Although the full environmental consequences of these systems are also not well understood, their required N use should be smaller than that of current first-generation crops.

Fig. 2. Estimated N deposition from global total N (NOy and NHx) emissions, totaling 105 Tg N y^{-1}. The unit scale is kg N ha^{-1} y^{-1}, modified from the original units (mg m^{-2} y^{-1}) (*16*).

increases in tropospheric O_3 can reduce C uptake in all systems (*29*). It remains a major research challenge to quantify all relevant N interactions sufficiently to estimate the net effect of Nr on climate forcing (*17, 26*).

How will tropical regions respond to rising N inputs? Much of our knowledge on N dynamics is from the temperate world, yet tropical regions will receive the most dramatic increases in Nr inputs over the next few decades [see, e.g., (*30*)]. Some tropical regions already experience elevated N deposition, acidic deposition, and aquatic eutrophication (*31*), both from urban development and from a combination of agricultural extensification and intensification. The trop-

world's dietary protein now comes from synthetic fertilizers, and estimates suggest that at least 2 billion people would not be alive today without the modern manifestations of the Haber-Bosch process (*21*). Yet, in many developed nations, the products from N-intensive agricultural practices lead to unhealthy diets, whereas elsewhere a lack of synthetic fertilizers, combined with depleted soil nutrient reserves, directly contributes to widespread malnutrition (*6*).

Once Nr enters the environment, its effects on terrestrial, aquatic, and atmospheric realms can influence human health and welfare in several ways. For example, N-driven increases in tropospheric O_3 pose direct health threats to humans

A Strategy for Now

There is compelling evidence that human alteration of the N cycle is negatively affecting human and ecosystem health. As demands for food and energy continue to increase, both the amount of Nr created and the magnitude of the consequences will also increase. Given the complexities of Nr use, its environmental mobility, and differences among regions, no single strategy will suffice (*40*). However, in keeping with the Nanjing Declaration on Nitrogen Management (*41*), here we highlight the intervention points in the global N cycle where N flows are concentrated and should be easiest to target (Fig. 3). We also give rough estimates of the decreases in Nr use or loss to the environment that are possible to achieve once the suggested strategies are implemented.

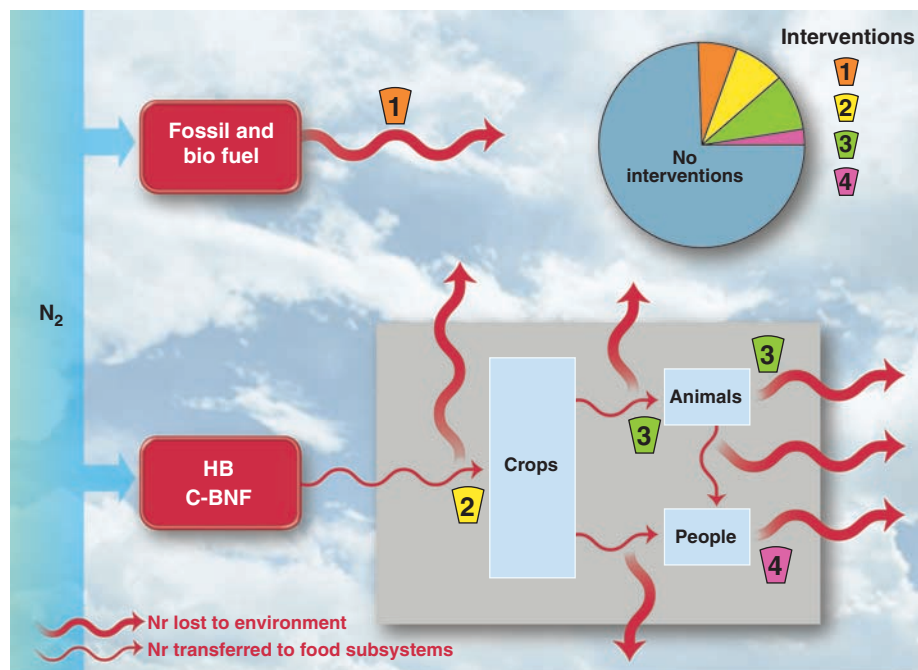Although we realize that the implementation will take time, the estimates are what might

Fig. 3. Conceptual model of where interventions in the N cycle can be used to decrease the amount of Nr created or the amount of Nr lost to the environment. The red boxes represent subsystems where Nr is created. The sky-background space represents the environment. Arrows leaving the red boxes either result in Nr lost to environment (fossil fuel and biofuel combustion) or inputs to the food production system (gray box). The light blue boxes within the gray box represent subsystems within the food production system where Nr is used. Nr can either enter these subsystems (thin red lines), or be lost to the environment (thick red lines). The numbers represent intervention points for N management. The pie chart shows the magnitude of Nr managed by the four interventions relative to the total amount created (187 Tg N) in 2005.

reasonably be expected to occur with current technology (see SOM text). First, controlling NOx emissions from fossil-fuel combustion using maximum feasible reductions would result in a decrease of Nr creation from 25 Tg N yr^{-1} to 7 Tg N yr^{-1}. Second, increasing nitrogen-uptake efficiency of crops would decrease Nr creation by about 15 Tg N yr^{-1}. Third, improved animal-management strategies would decrease Nr creation by about ~15 Tg N yr^{-1}. Fourth, even if only half the 3.2 billion people living in cities had access to sewage treatment, 5 Tg N yr^{-1} could be converted to N$_2$.

Together, these interventions represent a potential decrease of ~53 Tg N yr^{-1} created per year, or ~28% of the total Nr created in 2005. With this reduction, we would be able to largely offset the increases in Nr losses required for future growth in food, feed, fuel, and fiber production and energy use. Other intervention points are clearly needed if Nr creation rates are to decrease in the future. Although these estimates are necessarily rough, and implementing them would not be trivial, they indicate that a multipronged, integrated approach can decrease the amount of Nr lost to the environment.

Multiple comprehensive analyses of management strategies for some or all of these points have been made in recent years. Common to nearly all such analyses is a clear message that no single strategy will work.

We conclude by stressing two points. First, although reducing Nr creation and its unwanted impacts will be challenging, it is both possible and of critical importance. Second, not all management priorities are about reduction of Nr. Substantial and sustained intervention is also needed in regions that do not have sufficient Nr or other nutrients to sustain the population (8). In such regions, it will be important to seek ways to increase food production while minimizing nutrient loss and its subsequent environmental damages.

References and Notes

1. The term reactive nitrogen (Nr) as used in this paper includes all biologically active, photochemically reactive, and radiatively active N compounds in the atmosphere and biosphere of Earth. Thus, Nr includes inorganic reduced forms of N (e.g., NH$_3$ and NH$_4^+$), inorganic oxidized forms (e.g., NOx, HNO$_3$, N$_2$O, and NO$_3^-$), and organic compounds (e.g., urea, amines, and proteins), by contrast to unreactive N$_2$ gas.
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Supporting Online Material for

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SOM Text
References

Expansion of Discussion on Interactions of N and C in Terrestrial Ecosystems

The latest IPCC report *(S1)* undertook the most comprehensive assessment of Nr-climate interactions to date, but several critical components remain uncertain. The potential response of the C cycle offers a good example. Although earlier syntheses based on ^{15}N additions *(S2)* and modeled extensions of N budgets had suggested only a modest effect of N deposition on net forest C uptake, a recent synthesis of eddy flux data from northern hemisphere forests concluded that elevated N deposition is creating a sizable C sink *(S3)*. This finding is currently a matter of hot debate, as a major synthesis of European forest inventory data *(S4)* and a re-analysis of the Magnani et al. dataset *(S5)* both imply a C:N response that is an order of magnitude smaller than that claimed. In addition, even if an N-stimulated C sink is large, its future persistence is debatable, both because of progressive alleviation of N limitation, and because future increases in N deposition will be most pronounced in equatorial regions where N limitation is thought to be less widespread *(S6)*.

Background Details on Use of Interventions to Decrease Nr Loss to Environment

This material expands on the four intervention points mentioned in the paper's text and identified in Figure 3.

The first intervention point involves fossil fuel combustion. Using maximum feasible reductions, Nr creation during fossil fuel combustion can be decreased by about 30% of its current level (*S7*). The barriers to such a decrease are primarily financial, as the scientific, engineering and policy instruments are well developed. Utilizing this available technology, Nr creation from fossil fuel combustion would decrease from ~25 Tg N yr^{-1} to ~7 Tg N yr^{-1}.

The second intervention point is to increase the N use efficiency (NUE) of crop production. Cassman and colleagues estimate that there will be a 38% increase in global cereal demand by 2025. Compared to continuation of current trends, they project that this demand can be met with a 30% decrease in N fertilizer application to cereals if the decline in cereal harvest area (-0.33% yr^{-1} over 20 years) is halted and the crop yield response to applied N is increased by 20% *(S8)*. This 'intervention' would reduce the global Nr creation rate by ~15 Tg N yr^{-1}

Third, improving the genetic potential of the herd, animal feeding strategies and manure management would decrease N losses to the environment. It is estimated that improved animal feeding strategies in EU-27 could decrease by 2020 the total N excretion by ~13% and total N losses from animal manure by ~11%. A combination of

improved animal feeding and low-emission techniques could decrease by 2020 total N losses from animal manure in EU-27 by ~30%. Extending this to the world while considering the diversity in herd management, animal feeding and animal manure management, would result in a decrease of ~10-20 Tg lost to the environment (*S9*). Animal production systems are highly conducive to releasing Nr to the environment, so eating less meat would both substantially decrease Nr losses while having potential health benefits (*S10*)

The fourth intervention point involves sewage treatment. Each year, the global population produces ~20 Tg N in human waste of which <1% undergoes treatment that will convert it to N_2 (generally tertiary treatment). Even if only half of the 3.2 billion people who live in an urban environment had access to the required level of sewage treatment, this would result in 5 Tg N being converted to N_2.

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