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# HUMAN ALTERATION OF THE GLOBAL NITROGEN CYCLE: SOURCES AND CONSEQUENCES

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*Abstract.* Nitrogen is a key element controlling the species composition, diversity, dynamics, and functioning of many terrestrial, freshwater, and marine ecosystems. Many of the original plant species living in these ecosystems are adapted to, and function optimally in, soils and solutions with low levels of available nitrogen. The growth and dynamics of herbivore populations, and ultimately those of their predators, also are affected by N. Agriculture, combustion of fossil fuels, and other human activities have altered the global cycle of N substantially, generally increasing both the availability and the mobility of N over large regions of Earth. The mobility of N means that while most deliberate applications of N occur locally, their influence spreads regionally and even globally. Moreover, many of the mobile forms of N themselves have environmental consequences. Although most nitrogen inputs serve human needs such as agricultural production, their environmental consequences are serious and long term.

Based on our review of available scientific evidence, we are certain that human alterations of the nitrogen cycle have:

1) approximately doubled the rate of nitrogen input into the terrestrial nitrogen cycle, with these rates still increasing;

2) increased concentrations of the potent greenhouse gas $N_2O$ globally, and increased concentrations of other oxides of nitrogen that drive the formation of photochemical smog over large regions of Earth;

3) caused losses of soil nutrients, such as calcium and potassium, that are essential for the long-term maintenance of soil fertility;

4) contributed substantially to the acidification of soils, streams, and lakes in several regions; and

5) greatly increased the transfer of nitrogen through rivers to estuaries and coastal oceans.

In addition, based on our review of available scientific evidence we are confident that human alterations of the nitrogen cycle have:

6) increased the quantity of organic carbon stored within terrestrial ecosystems;

7) accelerated losses of biological diversity, especially losses of plants adapted to efficient use of nitrogen, and losses of the animals and microorganisms that depend on them; and

8) caused changes in the composition and functioning of estuarine and nearshore ecosystems, and contributed to long-term declines in coastal marine fisheries.

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## Introduction

The productivity and dynamics of many unmanaged terrestrial and marine ecosystems, and most agricultural and managed-forestry ecosystems, are limited by the supply of biologically available nitrogen. Humans are altering the global cycle of N via combustion of fossil fuels, production of nitrogen fertilizers, cultivation of nitrogen-fixing legumes, and other actions (Galloway et al. 1995). Increased N availability increases productivity and biomass accumulation substantially, at least in the short-term (Vitousek and Howarth 1991). Consequently, changes in N can alter the global cycle of C, affecting both the rate of increase of carbon dioxide in the atmosphere and the response of ecosystems to that increase (Schimel et al. 1995). Increasing N availability also generally reduces the biological diversity of affected ecosystems, and changes the rates and pathways of N cycling and loss (Tilman 1987, Berendse et al. 1993, Aber et al. 1995). Nitrate leaches through soils to stream water and groundwater, depleting soil minerals, acidifying soils, and altering downstream freshwater and coastal marine ecosystems (Likens et al. 1996, Nixon et al 1996); reactive oxides of N are important precursors of both acid rain and photochemical smog, and can be transported hundreds of kilometers to downwind ecosystems (Chameides et al. 1994); long-lived nitrous oxide contributes to anthropogenic enhancement of the greenhouse effect (Albritton et al. 1995). This report reviews and summarizes the extent of human alteration of the N cycle, and consequences for the functioning of terrestrial, freshwater, and marine ecosystems. It is not an exhaustive compilation of such studies—that would require volumes. Rather, it presents an overview of the current state of scientific understanding of this human-caused global change.

## Human Alteration of the Global N Cycle

The cycle of N is unique in that it consists of a massive, well-mixed, and (to most organisms) wholly unavailable pool of nitrogen gas ($N_2$) in the atmosphere; a relatively small and almost wholly biologically mediated conversion of $N_2$ to chemical forms of N that are available to most organisms; and a pool of N that cycles among plants, animals, microorganisms, soils, solutions, and sediments, and between land, water, and the atmosphere (Delwiche 1970). The most fundamental human-caused change to the global N cycle is a doubling of the transfer from the vast and unreactive atmospheric pool to biologically available forms on land (termed ''N fixation'').

As with any other human-caused global change, it is necessary to evaluate the state of the N cycle prior to extensive human alteration, as well as the magnitude of current human effects upon the cycle. And as with many global changes, determining the background state of the N cycle is difficult. Two natural processes transfer N from $N_2$ to biologically available forms—lightning and biological N fixation. The latter is carried out by microorganisms, many of them in symbiotic relationships with higher plants (especially legumes) and algae. In analyzing the global N cycle, the standard unit of measure is the teragram ($10^{12}$ g, abbreviated Tg), or million ($10^6$) metric tons of N. Lightning fixes $<10$ Tg N/yr now (Galloway et al 1995), and it has not been affected by human activity. Estimates of biological N fixation in marine ecosystems are variable and uncertain, ranging from $<30$ Tg/yr to $>300$ Tg/yr (Carpenter and Capone 1983, Carpenter and Romans 1991, Galloway et al 1995). Estimates of nitrogen fixation in terrestial ecosystems are better constrained; prior to extensive human activity, organisms probably fixed between 90 and 140 Tg N/yr (Soderlund and Rosswall 1982, Paul and Clark 1989, Schlesinger 1991). Several recent reviews demonstrate that human activity clearly has enhanced rates of N fixation on land substantially (Fig. 1) (Smil 1990, 1991, Vitousek and Matson 1993, Ayers et al 1994, Galloway et al 1995). A number of pathways are involved, including industrial fixation of $N_2$ for use as fertilizer, cultivation of crops with the capacity to fix N symbiotically, and mobilization and fixation during fossil-fuel combustion.

### *Sources of change*

*N fertilizer.*—Current industrial fixation of N for use as fertilizer totals $\approx$80 Tg/yr (FAO 1993). This figure does not include manures and other organic N fertilizers; globally, these account for more N than does industrial fertilizer, but manure application represents recycling of already-fixed N rather than new fixation. Industrial N fixation has increased exponentially from near zero in the 1940s. Until the late 1970s, most industrial N fertilizer was applied in developed countries, but use there has stabilized while applications in developing countries have increased dramatically. The immediacy and rapidity of the recent increase in N fixation is difficult to overstate. For example, Kates et al. (1990) point out more than half of all the industrially fixed N applied in human history up to 1990 had been used since 1980 (Fig. 2). The momentum of human population growth and increasing urbanization ensure that industrial N fixation will continue at high rates for decades.

*Fossil fuel combustion.*—The burning of fossil fuels

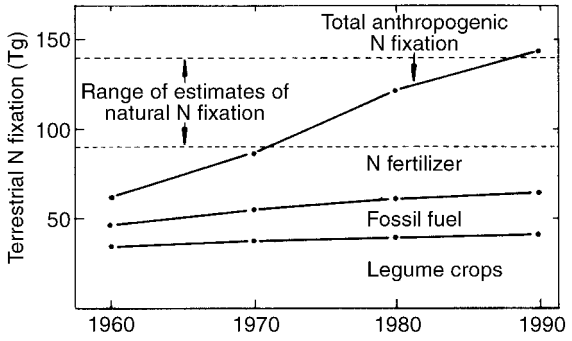


FIG. 1. Anthropogenic fixation of N in terrestrial ecosystems over time, in comparison with the range of estimates of natural biological N fixation on land. Modified from Galloway et al. (1995: Fig. 5).

transfers fixed N from long-term geological reservoirs to the atmosphere, and high-temperature combustion fixes a small amount of atmospheric $N_2$. A total of >20 Tg/yr of fixed N is emitted to the atmosphere during fossil-fuel combustion.

*Nitrogen-fixing crops.*—Leguminous crops and forages (i.e., soybeans, peas, alfalfa) support symbiotic N-fixing microorganisms, and thereby derive much of their N directly from atmospheric $N_2$. Fixation of N in excess of background rates in the natural communities that legume crops have replaced represents new, anthropogenic N fixation. There is also substantial biological N fixation associated with cultivation of some non-legumes, notably rice. The quantity of N fixed by crops is more difficult to determine than is industrial N fixation; Galloway et al. (1995) estimate it at 32–53 Tg/yr, and we will use 40 Tg/yr as an estimate here.

*Mobilization of N.*—In addition to enhancing fixation, human activity liberates N from long-term biological storage pools, and thereby contributes further to increasing the biological availability of N. The major pathways of mobilization are discussed in Vitousek and Matson (1993); they include biomass burning, which volatilizes >40 Tg/yr of N, with ≈20 Tg/yr of that fixed N (Lobert et al. 1990, Andreae 1993); land clearing and conversion, which could mobilize 20 Tg/yr; and the drainage of wetlands and consequent oxidation of their organic soils, which could mobilize 10 Tg/yr or more (Armentano 1980). Moreover, the loss of wetlands removes a significant sink for fixed nitrogen (denitrification, the conversion of nitrate to $N_2$ under anaerobic conditions), further increasing the mobility of N to and through streams and rivers (Leonardson 1994). All of these pathways have substantial uncertainties in both the quantity of N mobilized and its fate, but together they could contribute significantly to increasing the biological availability of N.

Overall, human activity causes the fixation of ≈140 Tg of new N per year in terrestrial ecosystems (Fig. 1)—at the upper end of the range of estimates for total background N fixation on land—and mobilizes perhaps 70 Tg more. It is fair to conclude that human activity has doubled (or more) the transfer of N from the atmosphere to biologically available pools on land. The added N is spread unevenly over Earth's surface—some areas (e.g., northern Europe) are profoundly altered (Berendse et al. 1993, Wright and van Breeman 1995), while others (e.g., remote south-temperate regions) receive little direct input (Galloway et al. 1982, Hedin et al. 1995)—but no place on Earth is unaffected. The recent increase in the quantity of fixed N in circulation is readily detectable in cores from the glacial ice of Greenland (Mayewski et al. 1986).

## EFFECTS ON THE ATMOSPHERE

The modern increase in fixation and mobilization of nitrogen is associated with increased emission, transport, reaction, and deposition of trace nitrogen gases, including nitrous oxide ($N_2O$), nitric oxide (NO), and ammonia ($NH_3$). Some human activities affect the atmosphere directly; for example, essentially all of the >20 Tg of N fixed or mobilized during fossil-fuel combustion and other high-temperature processes is emitted to the atmosphere as NO. Human activities also increase emissions indirectly. For example, agricultural fertilization increases the concentration of volatile $NH_3$ in soils, increases microbial processing of fixed N, and ultimately increases emissions of nitrogen gases from soils and groundwater (Eichner 1990, Schlesinger and Hartley 1992). Similarly, inadvertent N fertilization of unmanaged ecosystems downwind of agricultural/industrial areas can increase gas emissions from their soils.

These anthropogenic changes in the nitrogen cycle drive regional and global changes in the atmosphere. Nitrous oxide is increasing at the rate of 0.2–0.3%/yr, with most of the change occurring recently (Prinn et al. 1990). Nitrous oxide is a very effective greenhouse

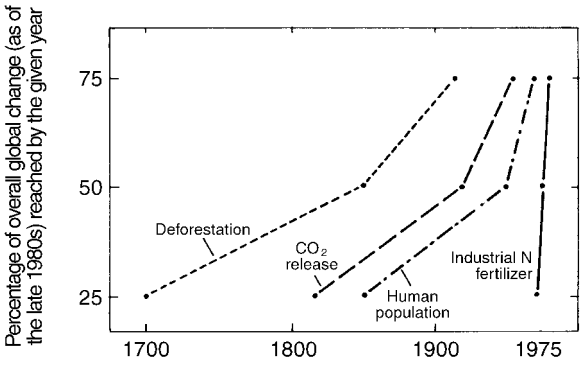


FIG. 2. Comparative timing of a number of global changes. Considering the extent of change as of the late 1980s as 100%, the figure shows the year by which 25%, 50%, and 75% of the overall change in deforestation, $CO_2$ release to the atmosphere, human population growth, and application of industrial fertilizer N had occurred. Revised from Kates et al. (1990:Fig. 1.1).

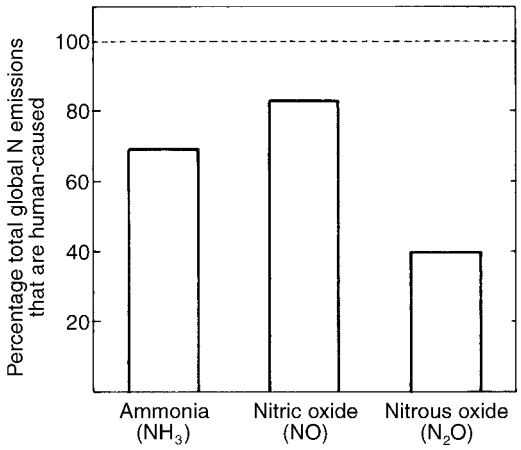


FIG. 3. The anthropogenic contribution to the total emissions of nitrogen-containing trace gases. Ammonia data are from Schlesinger and Hartley (1992), nitric oxide from Delmas et al. (*in press*), and nitrous oxide from Prather et al. (1995).

gas that absorbs infrared radiation in spectral windows not covered by other gases; it contributes a few percent to overall greenhouse warming (Albritton et al. 1995). It is unreactive in the troposphere, but it is destroyed by photolysis or by reaction with excited oxygen atoms in the stratosphere, where it can catalyze the destruction of stratospheric ozone (Crutzen and Ehhalt 1977). While the increasing concentration of $N_2O$ is clearly documented, the sources of that increase remain a matter of some discussion. Both fossil-fuel combustion and direct consequences of agricultural fertilization have been considered and rejected as the major source; there is a developing consensus that many anthropogenic sources (fertilizers, N-enriched groundwater, N-saturated forests, biomass burning, land clearing, nylon manufacture) all contribute to the increase (Prather et al. 1995). This ''dispersed source'' view is consistent with a terrestrial N cycle that has been systematically enriched by anthropogenic N fixation (Fig. 1).

In contrast to $N_2O$, NO and $NH_3$ are highly reactive in the atmosphere, and changes in their concentrations must be evaluated on local, regional, or subcontinental scales. Nitric oxide plays several critical roles in atmospheric chemistry. It affects the concentration of the main oxidizing agent in the atmosphere, the hydroxyl (OH) radical (Logan 1985). Moreover, it contributes (often in a rate-limiting way) to the photochemical formation of tropospheric ozone ($O_3$), the most important atmospheric gaseous pollutant in terms of its effects on human health and plant productivity (Reich and Amundson 1985, Chameides et al. 1994). When NO concentrations are high, the oxidation of carbon monoxide (CO), non-methane hydrocarbons, and methane ($CH_4$) leads to a net production of tropospheric ozone (Jacob and Wofsy 1990, Williams et al 1992); when NO concentrations are low, oxidation of these compounds is a sink for ozone. Finally, the end product of NO oxidation, nitric acid, is a principal component of acid rain. As with $N_2O$, a number of sources contribute to NO emissions, including microbial activity in fertilized soils. However, combustion is the dominant source; fossil fuel combustion emits >20 Tg/yr, and biomass burning (now mostly human-caused) may add about 8 Tg/yr more (Levy et al. 1991). Global NO emissions from soils total 5–20 Tg/yr (Yienger and Levy 1994, Davidson 1991) and a substantial fraction of this N is anthropogenic. Overall, 80% or more of all NO emissions globally are human-caused (Fig. 3; Delmas et al., *in press*).

Ammonia ($NH_3$) is the primary acid-neutralizing agent in the atmosphere, where it influences the pH of aerosols, cloudwater, and rainfall. As with NO, $NH_3$ emissions from ecosystems, transport in the atmosphere, and return to ecosystems via gas absorption, dry deposition, or in solution represent important pathways of nitrogen movement between ecosystems. Numerous studies have demonstrated substantial volatilization of fertilizer N as $NH_3$ (Fenn and Hossner 1985, Denmead 1990); Schlesinger and Hartley (1992) estimate $NH_3$-N fluxes from fertilized fields at 10 Tg/yr. Emissions from domestic animal wastes (32 Tg/yr) and biomass burning (5 Tg/yr) are also important globally; in sum, anthropogenic sources account for nearly 70% of all global ammonia emissions (Schlesinger and Hartley 1992; Fig. 3).

Enhanced emissions of N to the atmosphere have led to enhanced deposition of N on land and in the oceans. Based on extensive measurements of precipitation in remote areas of the southern hemisphere, where anthropogenic deposition of N is minimal, annual wet deposition of inorganic N in unpolluted regions averages 0.1–0.7 kg N/ha, of which 40% is nitrate and 60% is ammonium (Galloway et al. 1982, 1996, Likens et al. 1987). These fluxes are <10% of rates of wet deposition in the human-altered midwestern and eastern United States and <1% of rates in the most heavily affected areas of northern Europe (Berendse et al. 1993, Wright and Van Breeman 1995).

## EFFECTS ON TERRESTRIAL ECOSYSTEMS

### *N enrichment and the C cycle*

It is clear that rates of plant production and of the accumulation of biomass in whole ecosystems are limited by N supply over much of Earth's surface (Tamm 1991, Vitousek and Howarth 1991), particularly in temperate and boreal regions, and equally clear that human activity has increased N deposition substantially over much of this area. How much C is stored within terrestrial ecosystems as a consequence of anthropogenic N fixation and deposition? This question has important implications for the global cycle of C, in that deposition of anthropogenic fixed N could help to explain the ''missing sink,'' the imbalance between known $CO_2$

emissions from fossil fuel combustion and deforestation vs. known $CO_2$ accumulation in the atmosphere (Schimel et al. 1995).

Experimental work at European and American sites indicates that a large portion of the nitrogen retained by forest, wetland, and tundra ecosystems stimulates carbon uptake and storage (e.g., Rasmussen et al. 1993, Aber et al. 1995). Nitrogen deposition can also stimulate decomposition in some forests (Boxman et al. 1995), but the effects of added N in stimulating production are generally quantitatively more important (Hunt et al. 1988, Berg and Tamm 1991).

A number of analyses have calculated how much terrestrial C storage could result from N deposition (Peterson and Melillo 1985, Schindler and Bayley 1993, Hudson et al. 1994, Townsend et al. 1996); estimates of net C storage range from 0.1 to 1.3 Pg C/yr (1 Pg = 1000 Tg). The magnitude of potential C storage has tended to increase in more recent analyses, as the magnitude of change in the global N cycle is better appreciated (Keeling et al. 1996). The most recent analysis of the global C cycle by the Intergovernmental Panel on Climate Change (IPCC) concluded that N deposition could represent a major component of the missing C sink (Schimel et al. 1995). Further refinements could come from more complete analyses of the fraction of anthropogenic *N* that is retained within terrestrial ecosystems, on regional to continental scales (Galloway et al. 1995, Howarth et al. 1996).

### *Nitrogen saturation and ecosystem function*

Ultimately, there are declining returns in the response of plant production and carbon storage to additions of N, and consequently the potential for ecosystems to retain added N through increased production and organic matter storage is limited. The term ''nitrogen saturation'' (Ågren and Bosatta 1988, Aber et al. 1989, Aber 1992) has been applied to changes in N cycling in forest ecosystems that occur as N limitations to biological functions are relieved by N additions. In a fully N-saturated system (particularly one that is not storing C for some other reason such as its stage of stand development, or increasing $CO_2$), N losses to streams, groundwater, and the atmosphere should approach total N deposition, and any fertilization effects or continued C storage should disappear.

Concerns regarding the effects of N deposition on forest health and downstream ecosystems arose following observation of significant increases in nitrate concentrations in some lakes and streams (Grennfelt and Hultberg 1986, Henriksen and Brakke 1988, Stoddard 1994), and documented declines and mortality in coniferous evergreen forests in Europe (Schulze 1989). These observations have led to several field experiments that examined interactions between N deposition and forest ecosystem function (e.g., Van Miegrot et al. 1992, Kahl et al. 1993, Wright and van Breeman 1995, Magill et al. 1997). These experiments showed that where increased N additions led to increased nitrate mobility, the nitrate losses also led to losses of nutrient cations and increases in soil and water acidity (McNulty and Aber 1993, Boxman et al. 1995, Emmett et al. 1995).

Excess N availability also can cause nutrient imbalances in trees. These are expressed as root or foliar element ratios, especially Ca:Al and Mg:N ratios. As described below, Ca and Mg are lost via leaching, while the availability of Al is enhanced by acidity. Such imbalances may be linked to reductions in net photosynthesis, photosynthetic N-use efficiency, forest growth, and even increased tree mortality (Shortle and Smith 1988, Schulze 1989, Aber et al. 1995, Cronan and Grigal 1995).

In the northeastern United States, large shifts in foliar element ratios and increases in nitrate-leaching losses are generally restricted to high-elevation sites (which receive greater N deposition), those with shallow soils, those which have received little human disturbance (which presumably were close to input–output balance prior to receiving enhanced N deposition), and those receiving experimental additions of N well above ambient levels (e.g., Driscoll et al. 1987, Murdoch and Stoddard 1991, Kahl et al. 1993). In contrast, forests that have been subjected to intense or repeated biomass removals have very high capacities to retain N (Aber et al. 1995, Magill et al. 1997). The early stages of N saturation have also been noted in response to elevated N deposition in dry conifer forests surrounding the Los Angeles Basin, California (Bytnerowicz and Fenn 1996), in the Front Range of the Colorado Rockies (Baron et al. 1994), and in forests in which symbiotic N-fixing organisms are major components (Van Miegroet 1992). Nitrogen saturation is much further advanced over extensive areas of northern Europe, where rates of anthropogenic N deposition are several-fold greater than the most extreme areas in North America (Berendse et al. 1993).

Overall, the ability of a forest ecosystem to retain N is linked to its productive potential, and the degree to which previous disturbances have resulted in N removal and current N limitations. Thus, the extent and importance of N saturation are tightly linked to changes in land use, climate, atmospheric $CO_2$ and $O_3$, and other environmental variables that are also subject to rapid change.

### *Nitrogen deposition and changes in ecosystem composition and biodiversity*

The addition of limiting nutrients can dramatically change which species are dominant in ecosystems and markedly decrease the overall biodiversity of ecosystems. For example, experimental additions of nitrogen to grassland ecosystems in England have led to increased dominance by a few nitrogen-demanding grass

species, and to suppression of many other plant species (Lawes and Gilbert 1880, Brenchley and Warington 1958, Thurston 1969, Silvertown 1980). The highest rate of N loading caused the number of plant species to decline more than five-fold. Similarly dramatic reductions in plant diversity have been observed following N loading in North American grasslands (Tilman 1987, 1996, Huenneke et al. 1990), European grasslands (Bobbink et al. 1988), and European heathlands (Aerts and Berendse 1988).

Because of its high population density and interweaving of intensive livestock operations and industry, rates of N deposition in the Netherlands are the highest in the world, averaging 4–9 $g \cdot m^{-2} \cdot yr^{-1}$. The consequences of that enhanced deposition are well documented. Nitrogen deposition causes the conversion of heathlands to species-poor grasslands and forest (Aerts and Berendse 1988). Because heathlands occur on sandy, nitrogen-poor soils, the effect of deposition is to make heathlands more similar in composition to plant communities that occupy more fertile soils. Thus, biological diversity at the landscape level is reduced by N deposition, just as species richness within communities is reduced.

The loss of diversity caused by N deposition (or other global changes) can affect other aspects of ecosystem function. For example, recent experiments in the Midwestern United States showed that productivity in species-poor ecosystems that result from nitrogen addition was much less stable when they experienced a major drought (Tilman and Downing 1994). Similarly, the most diverse plots experienced much less year-to-year variation in productivity in response to climatic variation during non-drought years than did the most species-poor plots (Tilman 1996).

## Effects on Aquatic Systems

### *Historical changes in water chemistry*

Given human-caused acceleration of N fixation and other changes in N cycling, it is no surprise that N concentrations have increased over time in surface waters. In more-developed regions, nitrate concentrations have been more-or-less continuously measured in many rivers and other drinking-water supplies for decades. Analysis of these data show that nitrate has more than doubled in the Mississippi River since 1965 (Turner and Rabalais 1991, Justic et al. 1995), and that nitrate concentrations in major rivers in the northeastern U.S. have increased by 3- to 10-fold since the early 1900s (N. Jaworski and R. W. Howarth, *unpublished manuscript*) Available evidence suggests similar trends for many European rivers since the turn of the century (Paces 1982, Larsson et al. 1985), and Henriksen and Brakke (1988) report a doubling of nitrate in 1000 Norwegian lakes in less than a decade. Moreover, nitrate fluxes and concentrations in the large rivers of the world are correlated with human population densities

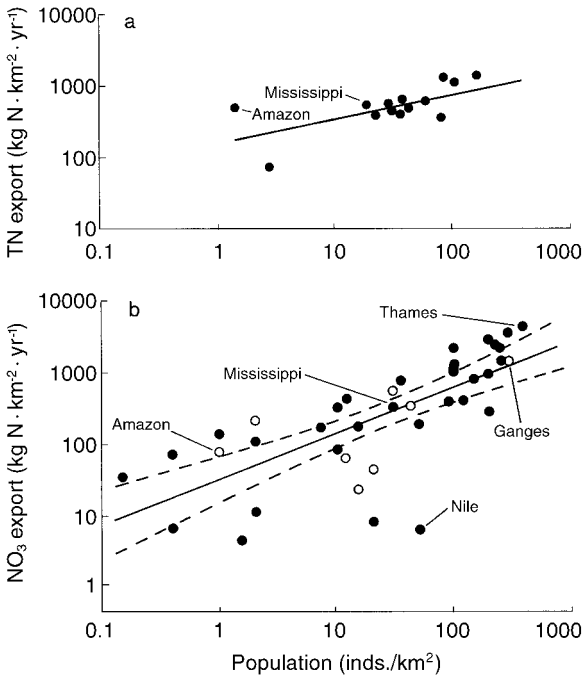


FIG. 4. Export of total nitrogen (a) and nitrate (b) from river systems, as a function of human population density in the watershed. Reprinted from Howarth et al. (1996:Fig. 3a, b) with permission of Kluwer Academic Publishers (where Fig. 3b was modified from Peierls et al. [1991]). Note logarithmic scale.

in the watersheds (Peierls et al. 1991, Cole et al. 1993) (Fig. 4).

No comparable historical data on the concentrations of total dissolved N in surface waters are available. Analyses of recent data suggest that N in streams and rivers draining relatively undisturbed forests is largely organic N (Schindler et al. 1980, Hedin et al. 1995); with increasing human disturbance, total N fluxes in rivers increase and a higher proportion is composed of nitrate (Howarth et al 1996). Total *N* fluxes in rivers also are correlated with human population density, but the slope is much shallower for total N than $NO_3$, illustrating the greater mobility of nitrate relative to other forms of N (Howarth et al 1996).

Fluxes of total N in temperate-zone rivers surrounding the North Atlantic Ocean are highly correlated with net anthropogenic inputs of N to their watersheds (Fig. 5; $r^2 = 0.73$, $P = 0.002$; Howarth et al. 1996). Net anthropogenic N inputs are defined as the sum of inputs as fertilizer, through N fixation by agricultural crops, as deposition of oxidized N from the atmosphere, and as the net import or export of N in food and feedstocks (see also Jordan and Weller 1996). For this regional analysis, N from sewage, N from animal feedlots, and $NH_4$ deposition from the atmosphere were considered to represent recycling of nitrogen within regions. For most of the regions surrounding the North Atlantic, anthropogenic inputs of N are dominated by fertilizer,

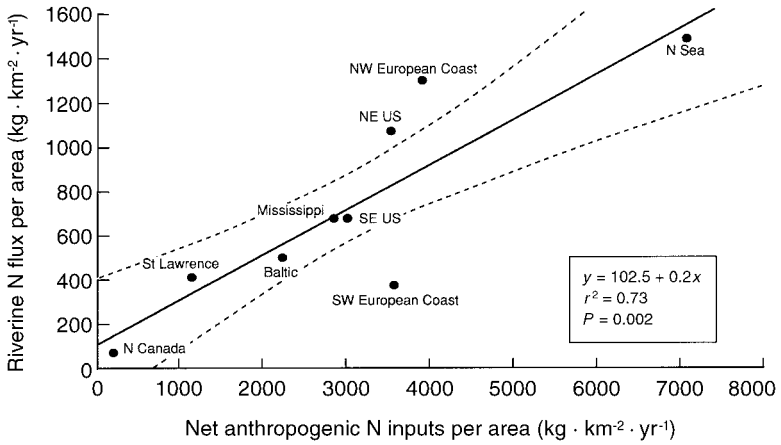


FIG. 5. Export of total nitrogen from watersheds surrounding the North Atlantic Ocean, as a function of net anthropogenic inputs of nitrogen to their watersheds. Net anthropogenic inputs are defined as (industrial N fertilizer + N fixation by legume crops + atmospheric inputs of oxidized N + net imports of N in food and feedstock). Reprinted from Howarth et al. (1996: Fig. 5a) with permission of Kluwer Academic Publishers.

but for the northeastern United States and the Saint Lawrence River and Great Lakes basin, atmospheric deposition of oxidized nitrogen is the greatest input (Fisher and Oppenheimer 1991, Howarth et al. 1996). Even though fertilizer inputs dominate in most regions, overall N exports from the regions are better correlated with atmospheric deposition of oxidized N.

Using relatively undisturbed areas as references, Howarth et al. (1996) estimated that riverine total N fluxes from most of the temperate regions surrounding the North Atlantic Ocean may have increased from preindustrial times by 2- to 20-fold. For the North Sea region, the N increase may have been 6- to 20-fold. Increased concentrations of nitrate have also been observed in groundwater in many agricultural regions (Moody 1990). The magnitude of this storage is difficult to determine, outside of a few well-characterized aquifers. Overall, the annual increment of N added to groundwater probably represents a small fraction of the increased nitrate transported in surface waters (Howarth et al. 1996), but the long residence time of groundwater in many aquifers means that decreases in groundwater quality are likely to continue as long as human effects on the N cycle are substantial.

Nitrate in drinking water represents a human health concern—when levels are high, microorganisms in the stomach may convert nitrate to nitrite. Nitrite absorbed into the bloodstream converts hemoglobin to methemoglobin, which is ineffective in oxygen transport in the blood. Acute and chronic elevated methemoglobin can kill infants, in a condition known as methemoglobinemia (Lee 1970). Although the disease is rare in the U.S., the potential for methemoglobinemia exists whenever nitrate levels exceed 10 mg N/L—the U.S. Public Health Service standard.

### *Nitrogen and acidification*

The interaction between nitrogen deposition and freshwater acidification is complex. In many areas, recent reductions in $SO_2$ emissions have reduced inputs of sulfuric acid to ecosystems, while emissions of the nitrogen oxides that are precursors of nitric acid have gone unchecked. As a result of this shift, and the fact that many catchments in moderate- to high-deposition areas appear to be becoming nitrogen saturated, nitric acid is playing an increasing role in lake acidification. In addition, nitric acid is highly mobile in snowpacks subjected to periodic melting, and in many areas it is the predominant strong acid in the spring acid pulse (Schaefer et al. 1990).

Nitrogen contributes to acidity in two ways—nitric acid does so directly, and ammonium deposited to terrestrial and aquatic systems can be a further source of acidity, in that both biological uptake of ammonium and nitrification produce hydrogen ions (Schindler et al. 1985, Schuurkes and Mosello 1988, Johnson et al. 1991). Moreover, where human alteration of the N cycle induces nitrate loss, the nitrate is a mobile anion that can move through soils to streams and groundwater, pulling cations with it and thereby depleting the soil of calcium and other nutrients (Likens et al. 1970, 1996). As calcium is depleted the leaching of toxic inorganic aluminum is increased. In N-saturated areas of Europe, a substantial fraction of atmospheric nitrate moves through terrestrial ecosystems without ever being taken up by organisms (Durka et al. 1994). The effects of acidity, and of the mobilization of aluminum that it causes, on aquatic systems have been reviewed by Irving (1990) and others. In areas without much acid-neutralizing capacity, they are clear and profound. The crucial point is that recent increases in the fixation and emission of N, and the increasing extent of N-saturated ecosystems, mean that controlling emissions of compounds that produce sulfuric acid will be insufficient to decrease acid rain or its effects on streams and lakes (Posch et al. 1995). The importance of N in acid-

ifying ecosystems is clearly recognized in Europe, where intergovernmental efforts are underway to reduce N emissions and deposition on a regional basis (DoE 1994). Critical loads (threshold levels) for nitrogen are considered to be an important part of managing acid precipitation there (Nilsson and Grennfelt 1988), and wetlands and riparian areas are being restored in an attempt to prevent excess nitrogen from entering freshwater and coastal zones (Leonardson 1994). An approach based on critical loads may not be appropriate conceptually for some of the consequences of N deposition—it is not clear there are thresholds for all of these effects—but critical loads may nonetheless represent a useful tool for managing N deposition.

Acidification itself can also disrupt the nitrogen cycle in freshwater ecosystems. In experimental lakes in Ontario, nitrification of ammonium ceased at pH values below 5.6 (Rudd et al. 1988), while higher nitrate inputs stimulated aquatic denitrification (Rudd et al. 1990). Nitrogen fixation in Little Rock Lake ceased at pH $<$ 5, but no effect on nitrification was observed (Schindler et al. 1991).

In freshwater ecosystems with sufficient phosphorus, additions of inorganic nitrogen can cause eutrophication; this can occur either independently or coupled to acidification (Schindler et al. 1985). Decreased diversity of both animal and plant species generally accompanies both eutrophication and acidification (Schindler 1990, 1994).

### *Fertilization and eutrophication in estuaries and coastal seas*

The eutrophication of estuaries and coastal seas is one of the best-documented and best-understood consequences of human-altered N cycling (Howarth 1988, NRC 1993, Justic et al. 1995, Nixon 1995, Nixon et al. 1996); it represents perhaps the greatest threat to the integrity of coastal ecosystems (NRC 1993, 1994). In most temperate-zone estuaries and coastal seas, net primary production and eutrophication are controlled by nitrogen inputs (Fig. 6; Boynton et al. 1982, D'Elia et al. 1986, Howarth et al. 1995, Nixon et al. 1996). Some areas are controlled by P rather than N, particularly tropical lagoons where adsorption onto carbonate sands provides a major sink for phosphate, and in some temperate estuaries and seas that receive extremely high loading of N (Howarth et al. 1995). Most often, however, eutrophication is caused by anthropogenic N loading—in sharp contrast to the majority of lakes in the temperate zone, where phosphorus is the element most limiting net primary production and controlling eutrophication (Schindler 1977).

Eutrophication has substantial effects on ecosystem function and composition in estuaries. It can cause anoxia (no oxygen) or hypoxia (low oxygen) in stratified waters, and both anoxia and hypoxia appear to be becoming more prevalent in many estuaries and coastal

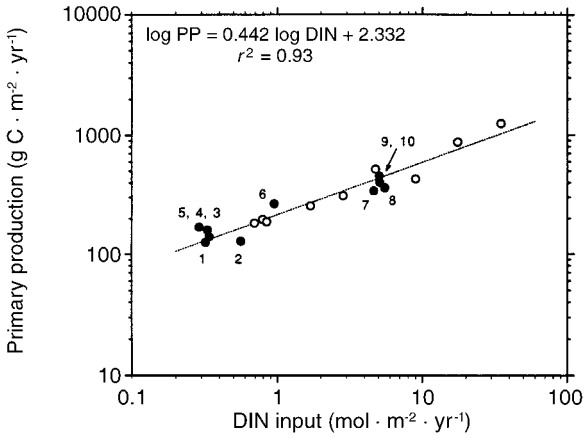


FIG. 6. Primary production (PP) by phytoplankton ($^{14}$C uptake) as a function of the estimated rate of input of dissolved inorganic nitrogen (DIN) per unit area in a variety of marine ecosystems. Natural systems are represented by solid circles; open circles are for large (13 m$^3$, 5 m deep), well-mixed mesocosm tanks at the Marine Ecosystems Research Laboratory during a multi-year fertilization experiment. Reprinted from Nixon et al. (1996:Fig. 5) with permission of Kluwer Academic Publishers.

seas (NRC 1993). There is good evidence for increasing anoxia since the 1950s or 1960s for the Baltic Sea (Larsson et al. 1985), the Black Sea (Lein and Ivanov 1992), and Chesapeake Bay (Officer et al. 1984). Hypoxic events have increased in Long Island Sound (Parker and O'Reilly 1991), the North Sea (Rosenberg 1985), and the Kattegat (Baden et al. 1990); low-oxygen problems have resulted in significant losses of fish and shellfish resources (Baden et al. 1990, Hansson and Rudstam 1990, NRC 1993).

Eutrophication is also associated with a loss of diversity, both in the benthic community and among planktonic organisms (Howarth 1991). Among phytoplankton, this can be manifested by dominance of nuisance algae. An increased incidence of nuisance algal blooms has been observed in many estuaries and coastal seas, and although the reasons are not completely known, compelling evidence suggests that increased nutrient supplies are responsible at least in part (Smayda 1989, NRC 1993). Toxic blooms of dinoflagellates (Anderson 1989, Burkholder et al. 1992) and of brown-tide organisms (Cosper et al. 1987) during the 1980s were responsible for extensive mortality of fish and shellfish in many estuaries. Eutrophication can also lead to a loss of diversity in subtidal beds of macroalgae, in seagrass beds, and in corals (NRC 1993).

## MAJOR UNCERTAINTIES

This analysis has focused on describing the scientific consensus on global change in the N cycle—in other words, what is known, and how we know it. There are also major uncertainties in our knowledge of the N cycle and how it relates to other aspects of global

change; we highlight a few of the most fundamental ones here. It is always possible, and often valuable, to improve estimates of a regional or global process; here, however, we focus on important processes that are known so poorly as to make it difficult to detect anthropogenic global change, or to predict its consequences.

### *Marine N fixation*

Credible estimates of the rate of N fixation in marine ecosystems range over more than a factor of 10 (Galloway et al. 1995); as a consequence, the state of scientific understanding of both current and background N fixation in the ocean is insufficient to evaluate the extent or global significance of any human-caused global change in marine N fixation. There is some evidence that human alteration of the N cycle could alter biological processes in the open ocean (Knap et al. 1986, Cornell et al. 1995, Michaels et al. 1996), but it is difficult to evaluate this possibility given our lack of understanding of the unmodified N cycle in the open oceans.

### *Changing resource limitation*

One consequence of human alteration of the global N cycle is that many ecosystems in which biological processes once were limited by N supply now receive large inputs of nitrogen, causing limitation by other resources to become more important. The dominant species in these systems may have evolved with nitrogen limitation; the ways they grow and function, and their symbiotic partners, could be highly tuned to it. How is the performance of organisms and ecosystems affected by shifts in resource limitation to conditions with which they have no evolutionary background, and to which they are not adapted?

### *Nitrogen retention capacity*

There is substantial variation in the capacity of forest ecosystems and wetlands to retain added N. A number of interacting factors that correlate with a system's capacity to retain N (prior to becoming N saturated) have been identified, including the C:N ratio of soil organic matter, soil texture and degree of chemical weathering, fire history, rate of biomass accumulation, and past human land use. Connections between ecosystems within landscapes can also affect losses of N to aquatic systems and the atmosphere. However, we lack a fundamental understanding of how and why the processes that retain N vary among systems, and how they have changed and will change with time.

### *Background N deposition and loss*

While information on current rates of N deposition and loss in developed regions is improving steadily, our understanding of these fluxes prior to extensive human effects is still patchy. In part, this reflects the fact that all of Earth is more or less affected by human activity. Nevertheless, studies in remote southern hemisphere temperate regions (Galloway et al. 1982, Hedin et al. 1995) illustrate that valuable information on areas that have been minimally altered by humans remains to be gathered.

### *Alteration of denitrification*

At the scale of large river basins, the majority of nitrogen inputs to a region probably are denitrified (Howarth et al. 1996). Our understanding of the locus of this denitrification is inadequate, although it is clear that riparian areas and wetlands make important contributions. Human activity has influenced the quantity and distribution of denitrification—enhancing it by adding nitrate to ecosystems, by building dams, and by cultivating rice; decreasing it by draining wetlands and altering riparian areas—but these changes remain poorly characterized.

## Future Prospects and Management Options

Globally, most of the anthropogenic enhancement of N fixation is closely tied to human activities related to food production. Intensive agricultural systems require large quantities of fixed N; humanity requires intensive agriculture to support our growing (and urbanizing) population; and our population is likely to double (or more) by the end of the next century. Moreover, N fertilizer is a relatively inexpensive commodity, and a decision to apply fertilizer is often the least expensive and most effective option to increase agricultural yield. The production and application of N has grown exponentially, and the highest rates of application are found in some developing countries with the highest rates of population growth (Matthews 1994). Galloway et al. (1994) suggest that by 2020 the global production of nitrogen fertilizer will increase to 134 Tg N/yr, from a current level of about 80 Tg N/yr. Clearly, curtailing anthropogenic fixation of N will be a very difficult challenge.

Nevertheless, there are prospects for slowing growth in the amount of N fixed for agriculture, and for reducing the mobility (and hence consequences) of the N that is applied. While the use of fixed N in agriculture cannot be substituted, there are reasons to believe that the efficiency of N fertilizer can be increased substantially. A relatively large fraction of applied N (often half or more) is typically lost from agricultural systems as $N_2$, trace gases, and nitrate; from a local viewpoint this is an expensive waste, while from a broader perspective it is a significant driver of global change. A number of management practices that can increase the efficiency of fertilizer N have been recognized. To the extent that these can be developed, improved, and implemented widely, some human fixation of N can be foregone. For example, Matson et al. (1996) evaluated N trace-gas flux in two commercial sugar cane plan-

tations in Hawaii. One applied N in several split applications, with increasing quantities of N timed to the requirements of the growing crop. Fertilizer was dissolved in irrigation water, which was delivered under the soil surface. The other plantation used fewer, larger applications of fertilizer N broadcast onto the soil surface. The more knowledge-intensive system used 2/3 as much N per crop as the more fertilizer-intensive system, and losses of $N_2O$ and NO (and probably other forms of N as well) were 10-fold less than from the fertilizer-intensive system. Moreover, the knowledge-intensive system yielded more and was more profitable; applying more knowledge proved cheaper than applying more fertilizer. The development and spread of this and similar technologies should be a high priority for ecologists as well as agronomists; they provide the opportunity to reduce costs and slow the rate of global change. The transfer of N-efficient technologies to developing regions is particularly crucial.

There are also ways that nitrogen lost from fertilized farmland can be prevented from reaching water courses, where it contributes to eutrophication. Such actions may be particularly important in areas adjacent to estuaries and other nitrogen-limited waters. For example, many streams have been channelized and wetlands drained to increase the area of agricultural land. We now realize that the elimination of wetlands and riparian areas has also eliminated important natural nitrogen traps, where much of the entering nitrate is denitrified. Restoration of such areas and the construction of artificial wetlands have been shown to be effective at reducing the transfer of nitrogen from agricultural land to the Baltic (Jansson et al. 1994 and other papers in *Ambio* 23, number 6).

The other major source of a human-fixed N is fossil-fuel combustion, which will also increase markedly, especially in the developing world, as we enter the next century. Galloway et al. (1994) suggest that the production of $NO_x$ from fossil fuels will be ≈46 TgN/yr in 2020, roughly double the current rate of emission. Improvements in the efficiency of fuel combustion and in the interception of its airborne byproducts must be implemented to reduce these emissions; again, it will be particularly important to transfer efficient technologies to developing economies.

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