

Human Contributions to Terrestrial Nitrogen Flux

Assessing the sources and fates of anthropogenic fixed nitrogen

Thomas E. Jordan and Donald E. Weller

Few species can assimilate the abundant dinitrogen gas in the atmosphere, so the supply of usable nitrogen compounds often limits biological production. The conversion of dinitrogen gas to biologically available forms of nitrogen is called nitrogen fixation. The global rate of nitrogen fixation has roughly doubled in the last few decades as a result of human activities (Galloway et al. 1995). Approximately 57% of anthropogenic nitrogen fixation comes from manufacturing fertilizer, 29% comes from cultivating nitrogen-fixing crops such as legumes, and 14% comes from burning fossil fuels (Galloway et al. 1995). The rates of these processes are increasing, but fertilizer production has increased the most. Worldwide fertilizer production has increased more than tenfold since the 1940s and continues to increase at a rate of approximately 5% per year (Galloway et al. 1995, Turner and Rabalais 1991). In addition to increasing food production, the enormous input of anthropogenic nitrogen has had many profound effects on the environment.

Most anthropogenic nitrogen

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Anthropogenic nitrogen from most sources comes together in the common pathways of the agricultural food chain

flows into and through the agricultural food chain (Figure 1). Newly fixed nitrogen is introduced to crops and pastures in fertilizer or by nitrogen-fixing bacteria associated with cultivated plants. Much of this nitrogen is incorporated into plants eaten by humans or livestock. Most of the nitrogen consumed by livestock goes into manure and urine, with some of the nitrogen in livestock waste being recycled to crops and pastures.

Atmospheric deposition of nitrate is another source of new available nitrogen to the biosphere (Figure 1). Nitrate deposition comes mostly from nitrogen oxides produced by burning fossil fuels (Berner and Berner 1987). In contrast, atmospheric deposition of ammonium is not a source of newly fixed nitrogen but rather a return of ammonia emitted mostly from livestock waste and fertilized soil (Schlesinger and Hartley 1992). Atmospheric deposition of anthropogenic nitrogen has stimulated growth in some forests (Schindler and Bayley 1993) but has reduced productivity in others (Aber

et al. 1989).

Some of the nitrogen in transit through crops, pastures, livestock, and people is carried away by flowing water (Figure 1). Nitrate leached from agricultural lands into groundwater has accumulated to toxic concentrations in many areas (Power and Schepers 1989). Export of nitrogen by streams increases as the proportion of agricultural land in the watershed increases (e.g., Rekolainen 1990). Recent increases in the discharge of nitrogen and phosphorus by the Mississippi River into the Gulf of Mexico have been linked to increases in fertilizer application (Turner and Rabalais 1991). Nitrogen transport in large rivers is greater for rivers draining more densely populated basins, possibly due to sewage inputs, agriculture sustaining the human population, or, more likely, both (Cole et al. 1993). However, even the nitrogen in sewage comes from nitrogen introduced into the agricultural food chain (Figure 1).

Worldwide increases in riverine discharges of fixed nitrogen contribute to eutrophication of coastal waters, where primary production is often nitrogen limited (Nixon 1995). For example, agriculture is the main source of nutrient discharge from the watershed of Chesapeake Bay (Fisher and Oppenheimer 1991), where enrichment with both nitrogen and phosphorus contributes to the problems of excessive plankton blooms and extensive reaches of hypoxic waters (Officer et al. 1984).

Denitrifying bacteria convert fixed

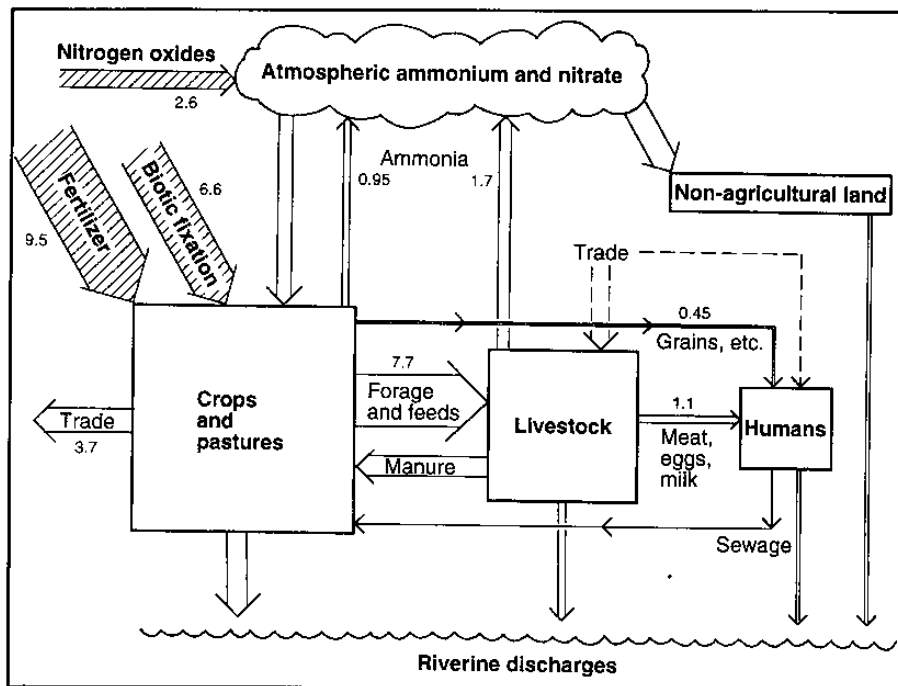


Figure 1. Major anthropogenic flows of nitrogen. Shaded arrows show inputs of newly fixed nitrogen through fertilizer application, biotic nitrogen fixation in agricultural lands, and production of nitrogen oxides that are converted to nitrate and deposited from the atmosphere. Unshaded arrows show other anthropogenic fluxes of nitrogen, including atmospheric deposition of ammonium and nitrate, emission of ammonia into the atmosphere, flows through the agricultural food chain, return of nitrogen to crops and pastures in animal wastes, and discharges of nitrogen in rivers. The widths of the arrows are proportional to the sizes of the flows for the entire coterminous United States. The numbers near arrows are our calculated estimates of flows (Tg nitrogen/yr). Dashed arrows show potential imports of nitrogen via trade in agricultural products, which are major sources of nitrogen in many regions, although the coterminous United States is a net exporter of nitrogen in agricultural products.

nitrogen back to dinitrogen, nitrous oxide, and nitric oxide gases. An increase in atmospheric nitrous oxide over the past few decades suggests that denitrification may have increased globally in response to the increase in anthropogenic nitrogen fixation (Prinn et al. 1990). Nitrous oxide contributes to global warming, which may perturb sea level, climate, precipitation patterns, and food production (Abrahamson 1989). Nitrous oxide also contributes to the depletion of stratospheric ozone that shields the earth's surface from ultraviolet radiation (Williams et al. 1992). Nitric oxide contributes to acid deposition (Williams et al. 1992).

At regional scales, the net trade of agricultural products can be an important source or sink for anthropogenic nitrogen because large amounts of nitrogen in agricultural products are transported over great distances

(Isermann 1993). However, such important interbasin fluxes of nitrogen carried by human transportation systems have been largely ignored by ecologists.

It is important to understand the fate of nitrogen that is fixed and transported by humans because of its potentially strong effects on terrestrial and coastal ecosystems. However, few complete large-scale analyses of anthropogenic nitrogen fluxes have been attempted. In this article we compare the nitrogen budgets for different regions of the United States, including all the major components of anthropogenic flux. We quantify the major sources of newly fixed nitrogen, fluxes through the agricultural food chain, and net exchanges of nitrogen in agricultural products. We test the hypotheses that riverine discharge of nitrogen increases as anthropogenic input of nitrogen increases and that the total net anthro-

pogenic input is a better predictor of riverine discharge than is any individual component of the anthropogenic input. Finally, to test the generality of our findings, we compare nitrogen budgets for regions of the United States with budgets for similar-sized regions in Western Europe, South America, and Western Africa.

Inputs of newly fixed nitrogen

For 15 major drainage basins of the coterminous United States (Figure 2), the sources and fates of newly fixed nitrogen were compared to riverine discharges of nitrogen from the same basins. Nitrogen fluxes were estimated for each county in the United States and summed according to drainage basin. For counties that fall into more than one basin, we apportioned the agricultural fluxes by the fraction of the county in each basin, which was determined by using a geographic information system to overlay the boundaries of basins (Seaber et al. 1987) and counties (USGS 1991). Nitrogen fluxes from the 15 basins were added to get total fluxes for the entire coterminous United States.

Data on nitrogen inputs from fertilizer in each county were from Alexander and Smith (1990).¹ Nitrogen fixation by crops and pastures was calculated by multiplying the areas of different types of crops and pastures in each county (Bureau of the Census 1993) by the following estimates of nitrogen fixation rates: soybeans, 78 kg nitrogen · ha⁻¹ · yr⁻¹; peanuts, 86 kg nitrogen · ha⁻¹ · yr⁻¹; nonlegume crops, 5 kg nitrogen · ha⁻¹ · yr⁻¹ (Barry et al. 1993, Messer and Brezonik 1983); alfalfa, 218 kg nitrogen · ha⁻¹ · yr⁻¹; and nonalfalfa hay, 116 kg nitrogen · ha⁻¹ · yr⁻¹ (Keeney 1979). The fixation rate used for dry edible beans (40 kg nitrogen · ha⁻¹ · yr⁻¹) was an average for cowpeas (Stevenson 1982), snap beans, and field beans (Keeney 1979). Eastern pastures, defined as any non-wooded pasture east of the Mississippi or within the Upper and Lower Mississippi regions (Figure 2), were assumed to fix 15 kg nitrogen · ha⁻¹ · yr⁻¹.

¹Also R. B. Alexander, 1994, personal communication. US Geological Survey, Reston, VA.

yr⁻¹ (Keeney 1979). Pastures or rangelands in other regions were assumed to fix 1 kg nitrogen · ha⁻¹ · yr⁻¹ (Woodmansee 1978). Atmospheric deposition of nitrate was calculated from measurements of wet deposition (NAPAP 1992), assuming that dry deposition of nitrate roughly equals wet deposition (Fisher and Oppenheimer 1991). Atmospheric deposition of ammonium was not considered to be a significant source of nitrogen because most of the ammonium deposited in a large drainage basin (Figure 2) is likely to come from ammonia volatilized within the basin (ApSimon et al. 1987).

Nitrogen fertilizer is the largest anthropogenic source of newly fixed nitrogen to the coterminous United States, but nitrogen fixation by pastures and cultivated plants is approximately two thirds as large (Figure 1). The largest estimated sources of biotic nitrogen fixation are alfalfa, soybeans, and nonalfalfa hay, which constitute 31%, 26%, and 24% of the total fixation by agricultural biota, respectively. Because nonalfalfa hay includes a variety of species, the nitrogen fixation rate for this category is uncertain. We used a fixation rate for hay in Wisconsin (Keeney 1979), but the rate is likely to vary with region and with the mixture of species. Pastures, rangelands, nonlegume crops, dry edible beans, and peanuts are relatively minor sources of biotic nitrogen fixation.

Atmospheric deposition of nitrate over the coterminous United States is only one-fourth as large as fertilizer input (Figure 1), but it is still an important source of newly fixed nitrogen, especially in the North-Central and Northeastern parts of the country, where deposition is highest. Unlike agricultural inputs, atmospheric deposition enters both non-agricultural and agricultural lands. Nitrate deposition directly on agricultural lands is approximately 1.2 Tg nitrogen/yr, or 7% of the total input of newly fixed nitrogen to agriculture in the United States. Adding nitrate deposition to agricultural lands, fertilizer application, and fixation by agricultural biota, we estimate that 17.3 Tg nitrogen/yr, more than 90% of newly fixed anthropogenic nitrogen, goes directly into croplands and pastures.



Figure 2. Major drainage basins of the United States. Basin boundaries correspond to US Geological Survey hydrological units (Smith et al. 1993) except that we have subdivided the South Atlantic-Gulf unit into two regions, Southeast and Eastern Gulf. Map is adapted from Smith et al. (1993).

Nitrogen flow in agriculture

We investigated the transport and transformation of nitrogen by agriculture to learn the fate of the anthropogenic nitrogen that directly enters the agricultural food chain. Shipment of agricultural products can be an important source or sink for anthropogenic nitrogen in many regions of the United States. Therefore, we calculated the net exchange of agricultural products for each county by summing the production of nitrogen as food, feed, and forage and subtracting nitrogen consumption by humans and livestock.

Calculating production of plant nitrogen. We calculated the amount of nitrogen in harvested crops from the total weights harvested in each county (Bureau of Census 1989) and the nitrogen content of the crops (Legg and Meisinger 1982). The nitrogen content of hay was assumed to be 1.5% (Messer and Brezonik 1983) rather than 2% (Legg and Meisinger 1982) because 1.5% produced more realistic estimates of grazing (described below). We did not individually quantify many of the various fruits and vegetables that humans eat. Instead, we estimated the total production of fruit and vegetable nitrogen from the areas of orchards and vegetable croplands (Bureau of the Census 1993) and the

rates of production of fruit nitrogen (35 kg · ha⁻¹ · yr⁻¹) and vegetable nitrogen (69 kg · ha⁻¹ · yr⁻¹) in Florida (Messer and Brezonik 1983). These rates may be too high for regions with shorter growing seasons, but they are reasonable for the Southeast and California, where fruit and vegetable harvests are most important.

We calculated the production of human and livestock foods separately by classifying crops according to whether their protein is fed to livestock or humans. For example, we classified sunflower seeds and cotton seeds as livestock feeds because, after their oils are extracted for human consumption, their protein-rich residues are mostly fed to animals. Soybeans are used similarly, but humans consume approximately 2% of the large harvest of soybean protein (Wedin et al. 1975). We classified sugar beets as livestock feed based on the fate of the nitrogen-containing residues after sugar extraction (Yamane 1982). Sorghum, hay, and corn grown for silage are clearly livestock feeds. Other crops were partitioned between livestock and human consumption based on Wedin et al.'s (1975) determination of the fraction of crops used for human food, industry (including alcohol), seed and feed together, and export. We simplified the analysis by assuming that protein residues from industrial use are fed to livestock and that the amount

Table 1. Production and fate of nitrogen (Tg/yr) in agricultural products of the coterminous United States.

Product	Yield	Loss in storage	US consumption	Export
Plant products:				
Concentrate feeds	7.4	0.74	3.6	3.1
Grazed nitrogen	2.5	0	2.5	0
Hay and silage	2.1	0.41	1.7	0
Plants for human foods	1.2	0.12	0.45	0.65
Total plant products	13	1.3	8.3	3.8
Animal products for humans	1.1	0.11	1.1	-0.09

used for seed is negligible. Thus, the proportion of nitrogen we attributed to human food was 61% for wheat, 17% for rye, 4% for corn, 6% for oats, and 3% for barley, with the remainder attributed to livestock feed. We also assumed that 50% of the peanut harvest is consumed by humans. We classified rice, dry edible beans, and potatoes as strictly human foods. We further assumed that exported commodities could be classified as food or feed in the same ratios as for domestic use.

Calculating consumption. Nitrogen eaten by livestock was calculated from the numbers of animals (Bureau of Census 1993) and their nitrogen consumption rates (Thomas and Gilliam 1977). We assumed that beef cows consume the amount given for 225–475-kg beef cattle in Thomas and Gilliam (1977), that heifers and heifer cows are equivalent to 113–224-kg beef cattle, and that other beef cattle are intermediate in their nitrogen consumption rate. We also assumed that pullet chicks and pullets consume the amount given for meat chickens in Thomas and Gilliam (1977). Production of nitrogen in human foods by livestock was calculated as percentages of the nitrogen eaten by livestock: 6.6% for beef cattle, 31% for dairy cattle, 27% for laying chickens, 18% for meat chickens and turkeys, 9.4% for hogs and pigs, and 5.7% for sheep (Pimentel et al. 1975). These production efficiencies apply to entire populations of livestock, including animals preserved for breeding (Pimentel et al. 1975). The nitrogen consumed by livestock but not incorporated into animal products was assumed to go to livestock wastes.

Consumption by sheep was assumed to equal manure production (SCS 1992) plus 5.7% that is meat produced for human consumption (Pimentel et al. 1975). Consumption by horses was assumed to equal manure production (SCS 1992). Human consumption of nitrogen was calculated by multiplying the number of people (Bureau of Census 1989) by the amounts of nitrogen consumed in animal- and plant-produced foods per person (4.4 and 1.9 kg nitrogen/yr, respectively, assuming protein is 16% nitrogen; Ehrlich et al. 1977). We assumed a 10% loss of all food and feed crops (except hay and silage) in storage and processing, based on the percentage of the cereal crop lost to pests in storage (Pimentel et al. 1975).

To estimate how much nitrogen was grazed by cattle, horses, and sheep from rangelands and pastures, we subtracted the nitrogen available in hay, silage, and protein concentrate feeds (e.g., grains and oil seed residues) from the total nitrogen demands of the grazers. We assumed that the hay and silage harvested within a county are consumed by grazing animals living within the county because there is little shipping of these bulky crops. We also assumed a 20% loss of hay and silage before consumption because these crops are handled and fed less efficiently than grains. We calculated the protein concentrates consumed by grazers using typical proportions of concentrates in their diets: 25% for beef cattle, 40% for dairy cows, and 11% for horses and sheep (Hodgson 1978, Wedin et al. 1975). The remaining nitrogen demand by grazers that is not satisfied by concentrates, hay, and silage was as-

sumed to be satisfied by grazing. However, we did not allow estimated grazing to exceed a maximum pasture production, which we calculated as the acreage of nonwoodland pastures times the production rate of hayfields per acre in the county. For counties in which pasture production could not satisfy the estimated grazing demand, we assumed that additional concentrate feeds were fed to the grazers.

Harvests and grazing. The first link in the agricultural food chain is the production of plant nitrogen for harvest or grazing. The crops yielding the biggest nitrogen harvests are soybeans and corn that is grown for grain. Together these crops account for almost half of the total nitrogen harvested and grazed in the coterminous United States. The third largest production of agricultural plant nitrogen is that grazed in pastures and rangelands. According to our estimates, grazed nitrogen represents 19% of the total production of plant nitrogen for harvest plus grazing (Table 1). Grazed plants are followed in nitrogen yield by wheat, alfalfa, and nonalfalfa hay (representing 8%, 7%, and 6% of the total, respectively). Humans eat approximately 9% of the total plant nitrogen produced, and most of the rest is eaten by livestock (Table 1).

The total amount of nitrogen harvested and grazed is approximately 77% of the 17.3 Tg/yr input of newly fixed nitrogen (including nitrate deposition) to agricultural lands. However, crops and pastures also receive nitrogen from livestock wastes and atmospheric deposition of ammonium, so the actual efficiency of converting total nitrogen input to consumable plant nitrogen is less than 77%. We do not know how much of the livestock wastes are applied to croplands and pastures, but it is likely to be a large proportion of the total. If all the livestock wastes were applied to agricultural lands, then the overall conversion efficiency of plants would be approximately 50%. Some livestock wastes are not applied to crops and pastures, so the actual efficiency is somewhere between 50% and 77%. This range is consistent with other analyses of nitrogen conversion effi-

ciencies for arable systems (Frissel 1977, Isermann 1993). Nitrogen inputs that are not recovered by harvests or grazing must be lost by leaching, runoff, denitrification, and ammonia volatilization because agricultural ecosystems usually do not accumulate much nitrogen in the soil (Frissel 1977). Some of the volatilized ammonia returns as atmospheric deposition on agricultural land, but the other losses remove nitrogen from the agricultural food chain.

Nitrogen flow through livestock. Most anthropogenic nitrogen in the United States is eventually consumed by livestock (Figure 1). Among livestock, beef cattle consume the most nitrogen, approximately 31% of the total plant nitrogen harvested and grazed. However, the beef cattle population of the United States produces much less food nitrogen for humans than does the dairy cattle population, the second most important nitrogen consumer. This is because a beef herd, including its breeding stock, converts only 6.6% of the nitrogen it consumes into food, whereas a dairy herd converts 31% of the nitrogen to food (Pimentel et al. 1975). Altogether, grazing animals in the United States, including cattle, horses, and sheep, obtain approximately 43% of their nitrogen from grazing, 27% from hay and silage, and 30% from grains and other protein concentrate feeds.

Nitrogen consumed by livestock but not converted to human food may still reenter the agricultural food chain as livestock waste applied to croplands and pastures. However, much livestock waste is lost through volatilization of ammonia, runoff, leaching, and denitrification from pastures, feedlots, and stored manure. Approximately 10%–40% of the nitrogen in livestock waste is released by volatilization of ammonia (ApSimon et al. 1987, Schlesinger and Hartley 1992). For the United States, a mid-range (25%) estimate of ammonia release from livestock wastes (1.7 Tg nitrogen/yr) exceeds our estimated incorporation of nitrogen in animal products for human consumption (1.1 Tg/yr). Another source of atmospheric ammonia is fertilizer. Approximately 10% (Schlesinger and Hartley 1992) of

Table 2. Areas, anthropogenic inputs of newly fixed nitrogen, net imports of food and feed nitrogen (negative numbers are exports), and average nitrate discharges from regions of the United States (Figure 2). Nitrate discharges are from Smith et al. (1993), except for the whole Mississippi basin, which is from Cole et al. (1993). Discharge from the coterminous United States is calculated as a weighted average of discharges from the individual regions. Discharge rates for Southeast and Eastern Gulf regions are each assumed to equal the discharge rate from their combined area given in Smith et al. (1993).

Region	kg nitrogen · yr ⁻¹ · ha ⁻¹ total area							
	Total area (10 ⁶ ha)	Newly fixed nitrogen					Total net input	Nitrate discharge
		Fixation by agricultural biota	Fertilizer	Nitrate deposition	Food and feed imports			
Individual regions:								
Northeast	42	8.3	6.0	6.8	10	31	2.0	
Southeast	32	6.0	12	5.0	4.6	27	0.79	
Eastern Gulf	36	5.7	13	5.0	5.8	29	0.79	
Great Lakes	30	15	18	6.8	-7.2	32	2.3	
Ohio-Tennessee	52	15	20	5.8	-11	30	3.0	
Upper Mississippi	48	27	40	5.4	-37	34	3.5	
Lower Mississippi	25	13	19	5.4	-13	24	1.2	
Missouri	130	12	13	2.8	-10	17	0.21	
Arkansas-White-Red	64	7.4	12	2.8	-0.65	21	0.20	
Souris-Red-Rainy	15	11	23	1.8	-19	17	0.039	
Texas	80	3.3	7.4	2.8	1.1	15	0.042	
Colorado	65	1.6	1.6	1.4	0.45	5.0	0.20	
Great Basin	36	2.4	0.86	0.9	0.49	4.6	0.17	
Northwest	70	3.8	6.4	1.4	-1.6	10	0.79	
California	41	3.4	12	1.4	3.5	20	0.17	
Aggregated regions:								
Whole Mississippi	334	14	19	3.8	-13	23	1.4	
Whole United States	767	8.7	12	3.4	-4.7	20	0.89	

fertilizer nitrogen (0.95 Tg nitrogen/yr) is volatilized as ammonia. Altogether, agriculture in the United States produces more atmospheric nitrogen in the form of ammonia (2.65 Tg/yr) than plant and animal nitrogen for human consumption (2.3 Tg/yr; Table 1). Globally, agriculture is the main source of ammonia emissions to the atmosphere (Schlesinger and Hartley 1992). Emissions of gaseous ammonia, unlike watershed discharges, can disperse agricultural nitrogen to systems that are uphill from source regions.

Net trade. Export of agricultural products is another major pathway of nitrogen flow. Assuming that production in excess of domestic consumption and losses is exported, we estimate that approximately 23% of the harvested and grazed plant nitrogen is exported from the United States as livestock feeds and approximately 5% as human food (Table 1). In contrast, there seems to be a small net import of nitrogen in animal products (Table 1).

Nitrogen flows in regions of the United States

Fertilizer and biotic fixation. There are dramatic regional differences in anthropogenic inputs of nitrogen that lead to regional differences in watershed discharges (Smith et al. 1993). Agricultural nitrogen inputs are highest in the Upper Mississippi region (Figure 2), in which a high proportion of the land is devoted to intensive corn and soybean farming. In this region, nitrogen inputs from fertilizer are 40 kg nitrogen · ha⁻¹ · yr⁻¹ (per total area of region including nonagricultural lands), and inputs from nitrogen-fixing crop and pasture plants are 27 kg nitrogen · ha⁻¹ · yr⁻¹ (Table 2). In contrast, the Great Basin region receives only 0.9 kg fertilizer nitrogen · ha⁻¹ · yr⁻¹, and 2.4 kg nitrogen · ha⁻¹ · yr⁻¹ is fixed by crop and range plants.

The relative importance of fertilizer and biotic nitrogen fixation differs among regions. Fertilizer inputs are approximately twice biotic fixation in four of the regions and 3.5

times fixation in the California region (Table 2). By contrast, biotic fixation is approximately equal to fertilizer input in the Colorado region and is three times fertilizer input in the Great Basin. Nitrogen fixation in pastures and rangelands accounts for less than 7% of the total fixation by agricultural biota in the United States, but it can be relatively more important in some regions. For example, pastures produce 28% of the biotic fixation in the Eastern Gulf region. However, our estimates of nitrogen fixation in pastures are uncertain because fixation rates may be affected by the mixture of plant species, by fertilizer application, and by grazing regime.

Nitrogen fixation in croplands and pastures is probably much greater than in nonagricultural lands. However, it is difficult to estimate fixation in nonagricultural lands because of the heterogeneity of natural ecosystems (Boring et al. 1988). Estimates of nitrogen fixation rates in forests range from 0.3–12 kg nitrogen · ha⁻¹ · yr⁻¹, with a median of approximately 1 kg nitrogen · ha⁻¹ · yr⁻¹ (Cushon and Feller 1989). Most of the nonagricultural land in the eastern United States is forested. If we therefore assume a fixation rate of 1 kg nitrogen · ha⁻¹ · yr⁻¹ for nonagricultural lands, then nitrogen fixation in nonagricultural lands would account for approximately 10% of the total biotic fixation in the Northeast, Southeast, and Eastern Gulf regions, but less than 5% of the total biotic fixation the Upper and Lower Mississippi, Ohio–Tennessee, and Great Lakes regions. In arid regions, nitrogen fixation in croplands and rangelands is low, so fixation in nonagricultural lands may be a larger fraction of the total. Estimates of nitrogen fixation rates in deserts range from 0.5 kg nitrogen · ha⁻¹ · yr⁻¹ (Wallace et al. 1978) to 25 kg nitrogen · ha⁻¹ · yr⁻¹ (Gist et al. 1978). Thus, nonagricultural lands could account for 10%–90% of the total biotic fixation in the Colorado and Great Basin regions.

Deposition. Atmospheric deposition of nitrogen in the form of nitrate is less than nitrogen fixation in croplands and pastures in all regions, and less than fertilizer inputs in all re-

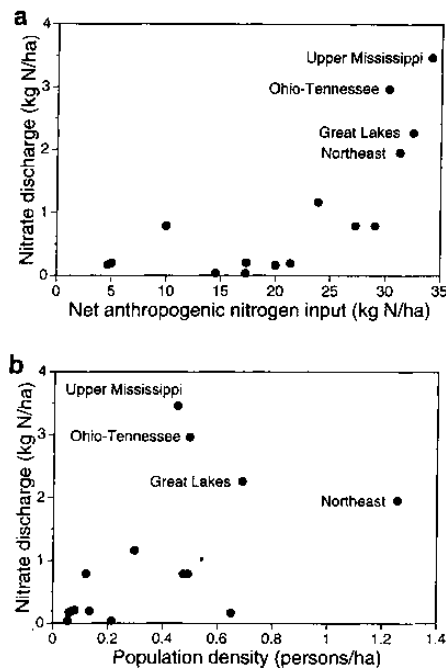


Figure 3. Annual nitrate discharge (Smith et al. 1993) versus annual net anthropogenic nitrogen input (a) and versus population (b) for major drainage basins of the United States (see Figure 2). The four basins with the highest nitrate discharges are labeled.

gions except the Northeast and the Great Basin (Table 2). Compared with agricultural inputs, nitrate deposition is most important in the Northeast region, where deposition rates are high and fertilizer inputs are moderate.

Plant production. Not surprisingly, the total yield of harvested and grazed plant nitrogen differs widely among regions. The total yield is highest in the Upper Mississippi region (67 kg nitrogen · ha⁻¹ · yr⁻¹), but the yield from grazing only is highest in the Arkansas–White–Red region (6.9 kg nitrogen · ha⁻¹ · yr⁻¹). The total yield of plant nitrogen in the Upper Mississippi region is approximately equal to the sum of biotic fixation and fertilizer inputs.

We estimated the harvest of nitrogen in most crops directly from agricultural statistics but estimated grazing with less certainty using assumptions about the diets of the grazers. In some regions, grazing is a large proportion of the total nitrogen yield from plants. For example,

in the Texas, Colorado, and Arkansas–White–Red regions, we estimate grazing to be 52%, 44%, and 40% of the total yield, respectively. Despite uncertainties, our grazing estimates exhibit realistic geographic patterns. Our estimates of production rates for grazing lands, averaged by region, ranged from 4–59 kg nitrogen · ha⁻¹ · yr⁻¹, with low productivity (less than 20 kg nitrogen · ha⁻¹ · yr⁻¹) in western regions and high productivity (more than 30 kg nitrogen · ha⁻¹ · yr⁻¹) in eastern regions. Regional differences in the estimated proportion of dietary nitrogen obtained by grazing are also realistic. Grazers in northern regions obtain less nitrogen from grazing than those in southern regions because in northern regions, feeding with hay supplants grazing in winter. Also, grazing is less important in regions such as the Great Lakes, with relatively high populations of dairy cattle, because dairy cattle consume higher proportions of feed concentrates (grain and oil seed cakes) than do other grazers (Hodgson 1978).

Net trade. Estimated net transport of nitrogen in agricultural products is often a large flux compared with inputs of newly fixed nitrogen (Table 2). In general, exports of agricultural products from a region tend to counterbalance inputs from fertilizer and biotic fixation. The greatest net import of nitrogen in agricultural products is 10 kg nitrogen · ha⁻¹ · yr⁻¹ to the Northeast region, where input from imported products exceeds any other nitrogen input. The greatest net export is 37 kg nitrogen · ha⁻¹ · yr⁻¹ from the Upper Mississippi region, where nitrogen export in the form of agricultural products exceeds biotic fixation and is slightly less than fertilizer inputs. Export of agricultural products from the Upper Mississippi region is a sink for 53% of the anthropogenic inputs of newly fixed nitrogen from fertilizer application, biotic nitrogen fixation, and nitrate deposition. For the entire Mississippi basin, export of agricultural products removes more than twice as much nitrogen (13 kg · ha⁻¹ · yr⁻¹; Table 2) as is removed in river discharge (5.7 kg · ha⁻¹ · yr⁻¹; Turner and Rabalais 1991).

Table 3. Correlations of average nitrate discharge with different variables for 12 regions of the United States, excluding 3 regions with low anthropogenic inputs (Northwest, Colorado, Great Basin). Correlations with $r^2 > 0.3$ are significant, with $P < 0.05$.

Independent variable	r^2
Net anthropogenic nitrogen input	0.76
Agricultural biotic nitrogen fixation	0.62
Atmospheric deposition of nitrate	0.61
Livestock waste nitrogen	0.44
Nitrogen fertilizer	0.36
Human population density	0.23
Import of agricultural-product nitrogen	0.22
Percent cropland in watershed	0.08

Riverine discharges

We compared our estimates of net anthropogenic nitrogen inputs to average discharges of nitrate from streams in each region reported by Smith et al. (1993). Nitrate discharge increases steeply as anthropogenic nitrogen input increases above 20 kg nitrogen \cdot ha $^{-1}$ \cdot yr $^{-1}$ (Figure 3a). When the input is 20 kg nitrogen \cdot ha $^{-1}$ \cdot yr $^{-1}$ or less, discharge is low, except for the Northwest region. The other regions may be able to absorb anthropogenic nitrogen inputs below 20 kg nitrogen \cdot ha $^{-1}$ \cdot yr $^{-1}$ without resultant increases in riverine discharges. Such a threshold for the effect of nitrogen input was not, however, observed in a similar analysis of riverine discharges of nitrate plus organic nitrogen (Howarth et al. in press).

Total net anthropogenic input is a better predictor of nitrate discharge than is any individual component of the input or any other variable that might influence the net input (Table 3). For example, human population density is a poor predictor of nitrate discharge ($r^2 = 0.23$; Figure 3b) compared with net anthropogenic nitrogen input ($r^2 = 0.76$; Figure 3a). The poor correlation with population density probably reflects the spatial separation of centers of human population from agricultural activities that strongly influence nitrate discharges. Thus, the highly populated Northeast region discharges only moderate amounts of nitrate, whereas the moderately populated Upper Mississippi region, with intense agriculture, discharges large amounts of nitrate (Figure 3b). In contrast, population density is a good predictor of

nitrate discharge from large river basins throughout the world (Cole et al. 1993), probably because net anthropogenic nitrogen input correlates with population density over the wide range of population density found among the world's large river basins. The percentage of cropland in a region is a poor predictor of nitrate discharge (Table 3), in contrast to findings of studies of smaller watersheds (e.g., Rekolainen 1990). Apparently, differences in farming practices among regions are too great to predict the effect of agriculture simply from the area of cropland. Instead, our analysis shows that regional nitrate discharge depends on the overall balance of anthropogenic nitrogen fluxes, and that neither the percentage of cropland nor the human population density provides an adequate measurement of that balance.

The observed correlation of riverine nitrate discharge with net anthropogenic nitrogen input (Figure 3a) is insensitive to uncertainties in estimating net input. To test the effects of uncertainties in our calculations, we first investigated which components of net nitrogen input have the greatest potential to change the strength of the correlation with riverine discharge. Using Monte Carlo simulations, we varied estimates of nitrogen flux in fertilizer application, nitrate deposition, biotic nitrogen fixation, crop yield, consumption by livestock and humans, and loss in transfer to consumers. Each estimate was varied

within $\pm 5\%$ of its mean value by the same percentage in all regions. All 49 flux estimates were varied simultaneously and independently for 10,000 trials.

In order of decreasing importance, the strength of the correlation was most sensitive to the fraction of harvest lost in transfer to consumers, the amount of fertilizer applied, and the harvests of corn for grain and soybeans. However, the correlation remained statistically significant despite varying loss in transfer from 0%–100% of the crop harvest, varying fertilizer application by a factor of 0.6 to 2, and varying harvests of corn or soybeans by a factor of 0 to 1.5. These ranges of variation include all reasonable values. The correlation between riverine nitrate discharge and net anthropogenic input was less sensitive to the other 45 components of our input calculation. Our sensitivity analysis suggests that better knowledge of how much nitrogen is lost in transfer to consumers would be most valuable in refining our correlation analysis. If loss in transfer is greater than our assumed value of 10% (Pimental et al. 1975), then the strength of association would be greater than our current estimate ($r^2 = 0.76$).

The correlation of nitrate discharge with anthropogenic nitrogen input is also quite insensitive to possible errors in nitrate discharge measurements. In 10,000 Monte Carlo simulations, we varied nitrate discharges from the 12 basins simultaneously and independently using

Table 4. Anthropogenic inputs of food and feed nitrogen and newly fixed nitrogen per total area of region (kg nitrogen \cdot yr $^{-1}$ \cdot ha $^{-1}$ total area), including biotic nitrogen fixation, fertilizer application, and atmospheric deposition of nitrate. For the European countries, agricultural nitrogen fixation and net import of feed are listed, and for the tropical regions total biotic nitrogen fixation (mostly nonagricultural) and net import of food plus feed are listed.

Region	Biotic fixation	Fertilizer application	Nitrate deposition	Food plus feed import	Total net input
Netherlands	2.8 ^a	140 ^a	18 ^b	97 ^a	260
Denmark	6.7 ^a	89 ^a	18 ^b	42 ^a	156
Great Britain	13 ^a	65 ^a	18 ^b	3.7 ^a	100
West Germany	5.8 ^a	61 ^a	14 ^c	23 ^a	100
West Africa ^d	21	0.12	2	0.16	23
Amazon ^e	20	—	2.4	—	22

^aIsermann 1993.

^bTietema and Verstraten 1991.

^cBredemeier et al. 1990.

^dRobertson and Rosswall 1986.

^eSalari et al. 1982.

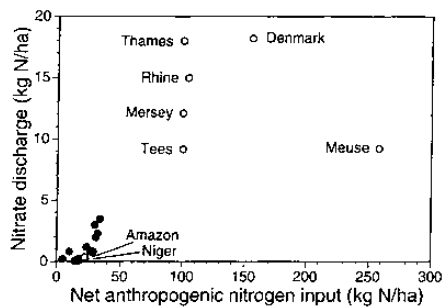


Figure 4. Annual nitrate discharge versus annual net anthropogenic nitrogen input (see Tables 2 and 4). Filled circles represent regions of the United States as in Figure 3. Open circles represent regions of similar size in other countries. The nitrate discharge data for Denmark is from Kronvang et al. (1993). Discharge data from the Amazon and Niger are from Cole et al. (1993). The discharge data from the Thames, Mersey, and Tees Rivers (Cole et al. 1993) are plotted against the input to Great Britain. The discharges from the Rhine and the Meuse Rivers (Cole et al. 1993) are plotted against the inputs to Western Germany and the Netherlands, respectively.

uniform random distributions from $\pm 50\%$ of the reported discharges (Smith et al. 1993). The strength of association varied from $r^2 = 0.39$ to $r^2 = 0.92$, and all the values within this range are statistically significant.

Anthropogenic nitrogen inputs, especially fertilizer application and feed import, are much higher in some Western European countries (Table 4) than in the subregions of the United States (Table 2), and nitrate discharges of rivers draining these countries are correspondingly high (Figure 4). In contrast, the anthropogenic inputs (Table 4) and nitrate discharges (Figure 4) of tropical regions in the Atlantic basin are low. In those regions, biotic nitrogen fixation is the major input. Although biotic fixation in the tropical regions is mostly nonagricultural, much of it occurs in young forests growing on lands abandoned from agriculture (Robertson and Rosswall 1986, Salati et al. 1982).

Riverine discharges of all forms of nitrogen combined, including nitrate and organic nitrogen, are generally smaller than net anthropogenic inputs of nitrogen. Total nitrogen discharges from the Mississippi basin

(Turner and Rabalais 1991) and the Northeast, Southeast, and Eastern Gulf regions (Howarth et al. in press) are approximately 25%, 35%, 25%, and 23%, respectively, of the net anthropogenic inputs (Table 2).

What is the fate of the remaining anthropogenic inputs? It is unlikely that much nitrogen is stored in biomass or soil in agricultural lands because agroecosystems seldom accumulate nitrogen (Frissel 1977). However, anthropogenic nitrogen dispersed via water-borne or atmospheric transport may accumulate as organic nitrogen in forest soil and wood. Forests in the United States (Turner et al. 1995) and worldwide are accumulating organic matter, possibly in response to increases in atmospheric carbon dioxide and to increases in deposition of nitrogen (Schlindler and Bayley 1993). Some anthropogenic nitrogen may also accumulate in groundwater. Many studies show trends in which the nitrate concentration of groundwater is increasing (Power and Schepers 1989), but it is not known how much nitrogen is accumulating in groundwater or how fast groundwater nitrogen is released to surface waters.

In addition, a large proportion of the nitrogen released from agricultural lands may be trapped or converted to gaseous forms in adjacent riparian zones or in other downstream aquatic ecosystems (e.g., Correll and Weller 1989). Much of the anthropogenic nitrogen may eventually be converted to gaseous forms through denitrification. Global increases in atmospheric nitrous oxide suggest that global increases in denitrification may parallel global increases in anthropogenic nitrogen fixation. However, denitrification rates for ecosystems are poorly quantified because of lack of accurate field methods and enormous spatial and temporal variability of denitrification rates (Weller et al. 1994). Thus, the fate of most anthropogenic nitrogen is unknown.

Conclusions

Humans have drastically altered the flow of nitrogen in the biosphere by nearly doubling the global fixation of nitrogen and by transporting agricultural products over long distances.

The resulting increase and redistribution of biologically available nitrogen is likely to have profound effects on ecosystems worldwide because nitrogen often limits plant growth. Also, increases in nitrogen availability may increase the global production of the atmospheric pollutants nitrous oxide and nitric oxide by denitrifying bacteria.

A complete understanding of human contributions to nitrogen flow in terrestrial systems requires an accounting of the three major anthropogenic sources of newly fixed nitrogen: application of nitrogen fertilizer, cultivation of nitrogen-fixing crops, and atmospheric deposition of nitrate. In addition to these sources, the net transport of nitrogen in agricultural products is an important source or sink for nitrogen in some regions. For example, import of agricultural products is the largest anthropogenic source of nitrogen to the northeastern United States, whereas export of products from the Mississippi basin removes more than twice as much nitrogen as is removed by riverine discharge from the basin.

Nitrogen inputs that are not removed by export of agricultural products can be dispersed to nonagricultural ecosystems through the air and water. For regions receiving more than $20 \text{ kg nitrogen} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$ of net anthropogenic nitrogen input, higher net inputs are associated with higher riverine discharges of nitrate. Increases in riverine discharges of nitrogen contribute to the eutrophication of coastal ecosystems worldwide. Riverine discharge is more closely correlated with the total net anthropogenic input than with any of the individual components of the input, probably because anthropogenic nitrogen from most sources comes together in the common pathways of the agricultural food chain.

Nitrogen is also dispersed through the air in ammonia emitted from fertilized soil and livestock waste. Agriculture in the United States releases more nitrogen as atmospheric ammonia than it incorporates into products for human consumption. Unlike riverine discharges, atmospheric emissions can disperse nitrogen to ecosystems uphill from source areas. However, emitted ammonia does not travel far before returning

to earth in wet or dry deposition. Therefore, there is little net transport of nitrogen as ammonia into or out of regions of the size we studied.

At most, approximately one third of the net anthropogenic nitrogen input to regions is discharged in rivers. Until we know the fate of the remaining two thirds of the anthropogenic nitrogen, we cannot know the full impacts of human alterations of nitrogen flux.

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