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Regional nitrogen budgets and riverine N & P fluxes for the drainages to the North Atlantic Ocean: Natural and human influences

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Abstract. We present estimates of total nitrogen and total phosphorus fluxes in rivers to the North Atlantic Ocean from 14 regions in North America, South America, Europe, and Africa which collectively comprise the drainage basins to the North Atlantic. The Amazon basin dominates the overall phosphorus flux and has the highest phosphorus flux per area. The total nitrogen flux from the Amazon is also large, contributing 3.3 Tg yr^{-1} out of a total for the entire North Atlantic region of 13.1 Tg yr^{-1}. On a per area basis, however, the largest nitrogen fluxes are found in the highly disturbed watersheds around the North Sea, in northwestern Europe, and in the northeastern U.S., all of which have riverine nitrogen fluxes greater than 1,000 kg N km^{-2} yr^{-1}.

Non-point sources of nitrogen dominate riverine fluxes to the coast in all regions. River fluxes of total nitrogen from the temperate regions of the North Atlantic basin are correlated with population density, as has been observed previously for fluxes of nitrate in the world's major rivers. However, more striking is a strong linear correlation between river fluxes of total nitrogen and the sum of anthropogenically-derived nitrogen inputs to the temperate regions (fertilizer application, human-induced increases in atmospheric deposition of oxidized forms of nitrogen, fixation by leguminous crops, and the import/export of nitrogen in agricultural products). On average, regional nitrogen fluxes in rivers are only 25% of these anthropogenically derived nitrogen inputs. Denitrification in wetlands and aquatic ecosystems is probably the dominant sink, with storage in forests perhaps also of importance. Storage of nitrogen in groundwater, although of importance in some localities, is a very small sink for nitrogen inputs in all regions. Agricultural sources of nitrogen dominate inputs in many regions, particularly the Mississippi basin and the North Sea drainages. Deposition of oxidized nitrogen, primarily of industrial origin, is the major control over river nitrogen export in some regions such as the northeastern U.S.

Using data from relatively pristine areas as an index of change, we estimate that riverine nitrogen fluxes in many of the temperate regions have increased from pre-industrial times by 2 to 20 fold, although some regions such as northern Canada are relatively unchanged. Fluxes from the most disturbed region, the North Sea drainages, have increased by 6 to 20 fold. Fluxes from the Amazon basin are also at least 2 to 5 fold greater than estimated fluxes from undisturbed temperate-zone regions, despite low population density and low inputs of anthropogenic nitrogen to the region. This suggests that natural riverine nitrogen fluxes in the tropics may be significantly greater than in the temperate zone. However, deforestation may be contributing to the tropical fluxes. In either case, projected increases in fertilizer use and atmospheric deposition in the coming decades are likely to cause dramatic increases in nitrogen loading to many tropical river systems.

Introduction

Human activity has greatly altered the nitrogen cycle on land, in aquatic systems, and in the atmosphere (Berner & Berner 1987; Galloway et al. 1995). Currently, global fixation of atmospheric N_2 for fertilizer, in combustion of fossil fuels, and by leguminous crops exceeds that by all natural sources, and changes in land use cause large additional amounts of nitrogen to be released from long-term reservoirs in both vegetation and soil (Vitousek 1994). These disturbances have been linked to a number of environmental concerns, including coastal eutrophication (Howarth 1988; Nixon 1992, 1995; National Research Council 1993; Gabric & Bell 1993; Paerl 1993; Justic et al. 1995; Rabalais et al., in press), acidification of freshwater lakes and streams (Driscoll et al. 1987; Henriksen & Brakke 1988; Kelly et al. 1990; Murdoch & Stoddard 1992), forest decline (Schulze 1989), climate change (Keller et al. 1986; Khalil & Rasmussen, 1992; Vitousek & Matson 1993), and shifts in community structure (Tilman 1984; Crabtree & Bazzaz 1993; Bowman et al. 1995) and in ecosystem function (McNulty et al. 1991; Aber et al. 1993; Burton et al. 1993; Neff et al. 1994). Accelerated nitrogen cycling has also been suggested to increase the sequestration of carbon in forests, slowing the rise of atmospheric carbon dioxide (Schindler & Bayley 1993). However, gaps in our understanding of nitrogen dynamics over large spatial scales often limit our ability to clearly identify the areas of greatest concern, and to predict the response of systems to future change.

The foundations of ecosystem ecology and biogeochemistry are based in part on studies of small-scale watersheds. Much less attention has been given to evaluating material fluxes through the landscape at larger scales, and we are only beginning to understand the rules that govern regional exchanges of nitrogen between atmospheric, terrestrial and aquatic systems. These exchanges may be especially crucial to the functioning of freshwater and coastal ecosystems, as even relatively small changes in the processing and retention of nitrogen applied to the terrestrial landscape could have a large impact on the downstream aquatic environment. In fact, several studies have noted much greater fluxes of nitrate in rivers over the past few decades. Nitrate concentrations in the Mississippi River have more than doubled since 1965 (Turner & Rabalais 1991; Justic et al. 1995; Rabalais et al., in press), and Pacés (1982) estimated that nitrate in many European rivers has probably increased 5- to 10-fold since the turn of the century. Such increases are presumably in response to increased use of nitrogen fertilizer, cultivation of nitrogen-fixing crops, greater population density, and increased atmospheric deposition of nitrogen. Total nitrogen fluxes have also probably increased, but historical data on total nitrogen in rivers are extremely rare.

Our overall objective in this study was to estimate total nitrogen export (TN) in all of the major rivers draining into the North Atlantic Ocean, to assess how much that export has changed since pre-industrial times, and to identify the principal agents of that change. The work presented here is part of a larger collaborative effort to assemble a nitrogen budget for all components of the North Atlantic region: atmosphere, watersheds, coastal systems, and the open ocean. Although the focus of the paper is on nitrogen, estimates of phosphorus (TP) are also developed because of the potentially valuable information provided by considering N:P ratios. In some cases, we have had to extrapolate estimates based on fluxes of organic carbon or other indirect approaches, but most of our estimates are derived from direct data on water fluxes and concentration of TN and TP (including dissolved and particulate inorganic and organic forms). These data are summarized by dividing the total study area into 14 major watershed regions. In addition to the estimates of TN and TP export by rivers, we present data on population density, sewage and wastewater inputs, fertilizer application, atmospheric deposition, fixation of nitrogen by leguminous crops, and the net import or export of nitrogen in agricultural products.

We chose the North Atlantic region for several reasons. One, when all components of the region are considered, it is probably the best studied area on the planet, and we therefore had the best chance to derive a reasonably accurate nitrogen budget for an entire ocean basin. In discussing transport of C, N, and P in the world's rivers, Meybeck (1982) notes that "it is very difficult to get information on more than half the river water discharging to the ocean" as a whole, and fewer data are available for TN than for C and P. However, for the North Atlantic Basin, we have direct data for TN fluxes in the majority of watersheds representing approximately 80% of the total freshwater discharge. Two, much of the region has been and continues to be significantly affected by human activity, and we wished to assess the impacts of such activity on regional-scale biogeochemistry. Finally, the region is comprised of a great diversity of environments, thereby allowing comparisons of many of the world's major biome types within the confines of our study boundaries.

Regional boundaries and data sources

We divide the watersheds which flow into the North Atlantic Ocean into 14 regions (Figure 1; Table 1). Seven of these regions are in North America, 2 are in Central and South America, 4 are in Europe, and one is in Africa. Regions are selected to coincide with discrete portions of the coastal ocean. The definition of the size and boundaries of regions is dictated in part by the availability of data and by previous studies. All estimates reported here are at the scale of these large regions, although estimates for many of the regions are calculated by summing fluxes for watersheds and smaller areas within the regions. Most nutrient flux estimates are for average periods during the 1980's, although estimates for a few regions are based on late 1970's data. We have explicitly not considered inputs from land into the Mediterranean Sea; net nitrogen fluxes between the Mediterranean and the Atlantic Ocean proper are addressed by the oceanic working group (Michaels et al., this volume).

North Canadian rivers

The "north Canadian rivers" region is defined to be all drainages into Hudson Bay plus the drainages which flow into the Atlantic from Labrador and Newfoundland; this is a large area which includes 40% of Canada (Laycock 1987). Data for area, population, and freshwater discharges are all from Laycock (1987). Annual fluxes of total nitrogen (TN) and total phosphorus (TP) from this region are estimated as the product of the average TN and TP concentrations in 6 rivers in the region (17 μM N and 0.44 μM P; Paré & Goulet 1980; Schetagne 1981; Schetagne & Roy 1985) and annual water discharge to Hudson Bay and coastal discharges of streams and rivers in Labrador and Newfoundland (Laycock 1987).

St. Lawrence

The "St. Lawrence" region includes the drainage basins of the Great Lakes (van der Leeden et al. 1990) plus areas of Canada labeled by Laycock (1987) as "St. John-St. Croix," "maritime coastal," and "North Shore-Gaspé." The area of this region is calculated from information in Laycock (1987) and van der Leeden (1975). Freshwater discharge is the sum of discharge for the St. Lawrence River (Pocklington & Tan 1987) and regional discharge from the

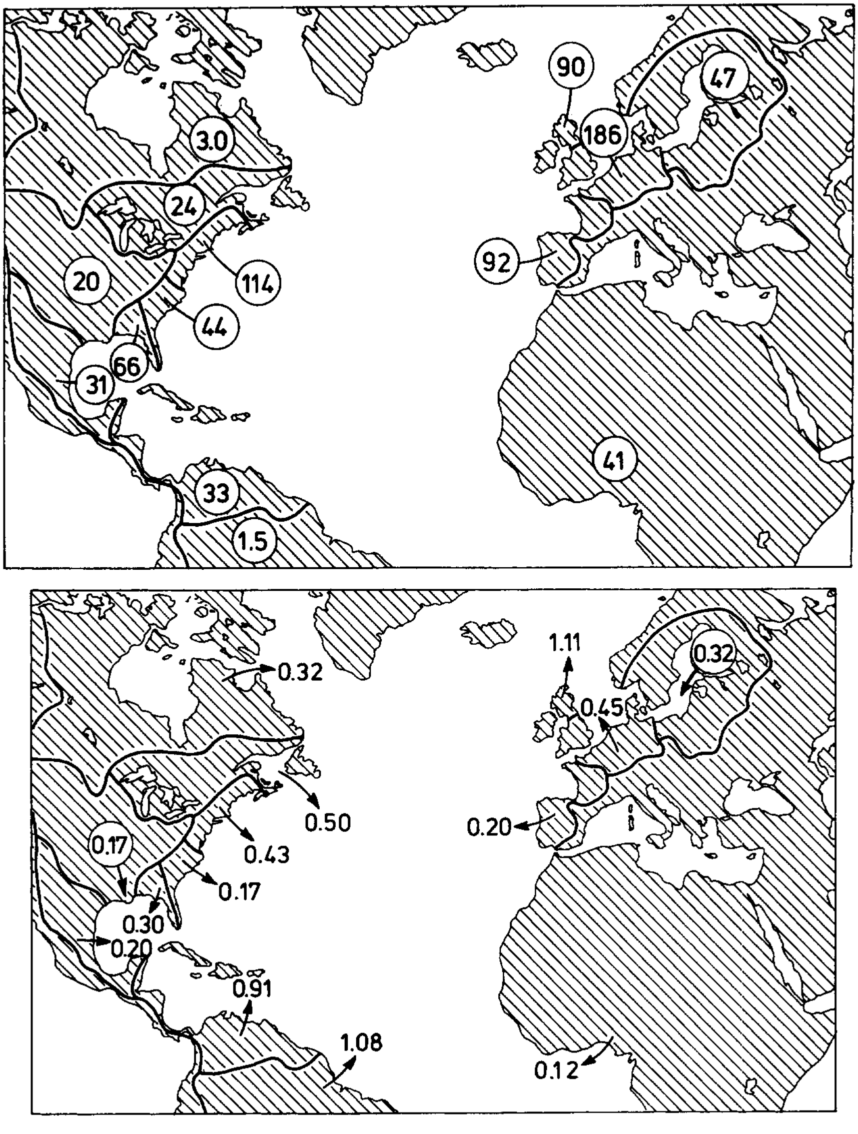

Figure 1. Population density of each of the 14 regions in individuals per km^2 (top, a), and water discharge per area in m^3 m^{-2} yr^{-1} (bottom, b).

St. John-St. Croix, maritime coastal, and North Shore-Gaspé areas (Laycock 1987). Population for the St. Lawrence region is estimated by summing the population of the St. John-St. Croix, maritime coastal, and North Shore-Gaspé areas (Laycock 1987) and the St. Lawrence and Great Lakes basins; population in the St. Lawrence and Great Lakes basins is the product of land

Table 1. Summary of regional data on drainage area, population, and water discharge.

	Drainage area (10^6 km^2)	Population (10^6 ind.)	Water discharge (10^9 m^3 yr^{-1})
North Canadian rivers	3.98	12	1,260
St. Lawrence basin	1.60	38	801
NE coast of US	0.48	55	208
SE coast of US	0.35	16	59
Eastern Gulf of Mexico	0.35	23	106
Mississippi basin	3.23	64	546
Western Gulf of Mexico	1.42	44	285
North America – total	11.4	252	3,260
Caribbean Islands and Central America (incl. Orinoco)	2.28	75	2,070
Amazon & Tocantins basin	6.49	10	7,000
Central & South America – total	8.77	85	9,070
Baltic Sea	1.50	70	475
North Sea	0.84	156	380
NW European coast	0.34	30	378
SW European coast	0.55	50	110
Western Europe – total	3.23	306	1,340
NW Africa	3.53	146	416
Total – all basins	26.9	790	14,100

Note: Watersheds flowing into Mediterranean Sea are not included.

area and mean population density in the states and provinces of this region (22 individuals km^{-2}; Espenshade 1990). Annual fluxes of TN from the region are taken as those of the St. Lawrence River at Quebec City (Comité d'étude sur le fleuve Saint-Laurent 1978) plus contributions from the North Shore-Gaspé, St. John-St. Croix, and Maritime Coastal regions calculated as the product of regional water discharges (Laycock 1987) and average nutrient concentrations. Annual fluxes of TP are estimates from a phosphorus budget for the upper estuary of the St. Lawrence (Lucotte 1989) plus contributions from the three coastal regions. Nutrient concentrations for these regions are assumed to be the same as in the North Canadian Rivers region.

NE coast of US

The NE coast of the US corresponds to a water resource region defined by the U.S. Geological Survey (Crippen 1969). Area and freshwater discharge are from Crippen (1969). Population data and estimates of TN and TP fluxes are from Jaworski and Howarth (in press, and ms. in prep.). Jaworski and Howarth (in press, and ms. in prep.) use many data sources but rely most heavily on data from characterization studies for individual estuaries in the region as part of the Natural Estuary Program of the U.S. Environmental Protection Agency and state environmental agencies.

SE coast of US

The SE Coast of the U.S. also corresponds to a U.S.G.S. water resource region (Crippen 1969). Area and freshwater discharge data are from Crippen (1969). The population is estimated from state population data (Espenshade 1990) using a delineation by Crippen (1969) for assignment of what percent of each state lives within the region. Annual flux of TN from the region is estimated as the sum of independent estimates for nitrate fluxes per area, dissolved organic nitrogen per area, and particulate organic nitrogen per area multiplied by the area of the drainage. The nitrate flux per area is from Smith et al. (1993). Dissolved and particulate organic nitrogen fluxes are estimated from dissolved and particulate organic carbon fluxes in the region (Degens et al. 1984, as cited in Meybeck et al. 1989) by assuming a C:N molar ratio of 23.3 for dissolved organic matter (Meybeck 1982) and of 9.7 for particulate organic matter (Ittekot & Zhang 1989). The flux of TP is estimated from Smith et al. (1993).

Eastern Gulf of Mexico

Boundaries are those defined by the U.S.G.S. water region (Crippen 1969). Area, freshwater discharge, population, and annual fluxes of TN and TP from the region are estimated as for the "SE coast of U.S." region using the same data sources. These two regions have much in common; we consider them separately since one drains into the Gulf of Mexico while the other drains into the North Atlantic Ocean proper.

Mississippi basin

This region is delineated by the natural drainage basin of the Mississippi River and its tributaries. It is a large region, making up 41% of the area of the lower 48 states of the United States (Wright 1993). Data on population, area, and freshwater discharge are from Meybeck et al. (1989). Annual fluxes of TN and TP are from Turner & Rabalais (1991).

Western Gulf of Mexico

The "western Gulf of Mexico" region includes another U.S.G.S. water resource region (Crippen 1969) but also watersheds within Mexico from the Rio Grande to the tip of the Yucatan which flow into the Gulf of Mexico (del Rio & Wilson 1969). This includes most of Mexico. Population in this region is given by the population of Texas (Espenshade 1990) plus the product of the drainage area within Mexico (del Rio & Wilson 1969) and the average population density of Mexico (Espenshade 1990). Regional discharge is calculated by summing the flows within the U.S. (Crippen 1969) and those within Mexican drainages to the Gulf (del Rio & Wilson 1969). Fluxes of TN and TP are estimated as for the "SE Coast of U.S.," using the same data sources. For TP and the nitrate portion of TN, values of Smith et al. (1993) for the portion of this region within the U.S. are extrapolated to the portion of the region within Mexico as well.

Caribbean Islands and Central America

This region extends from the Yucatan Peninsula to the French Guiana/Brazil border and includes all islands in the Caribbean Sea. Area in the region is the sum of the area of all of the major Caribbean islands (Espenshade 1990) and the area on the mainland, consisting of a Central American portion and a South American portion. The Central American area is estimated from the fractional area of the Central American isthmus which drains into the Caribbean (0.692; Ahlgren et al. 1969) multiplied by the area of Guatemala, Honduras, Nicaragua, Costa Rica, and Panama (Espenshade 1990). The amount of area in this region in South America is obtained by summing the principal drainage areas, including the Orinoco, the Magdalena, and the principal rivers of Surinam, Guyana, and French Guiana (van der Leeden 1975; Lewis & Saunders 1989). Population is the sum of the population of the Caribbean Islands (Espenshade 1990), the population of countries in the South American portion of this region (Colombia, Venezuela, Guyana, Surinam, French Guiana; Espenshade 1990), and population of the Central American countries in the region (Espenshade 1990) multiplied by the percentage area of these countries which flow to the Atlantic (Ahlgren et al. 1969).

Discharge data for the Central American area are from Ahlgren et al. (1969) and for the South American area are from van der Leeden (1975) and Lewis & Saunders (1989). Fluxes of TN and TP from the region are based on a discharge-based extrapolation of estimates for the Orinoco River (Lewis

& Saunders 1989) plus estimates for sewage and wastewater inputs. The Orinoco is the largest river in the region and contributes 55% of total freshwater discharge for the region. Sewage and wastewater inputs are added to this extrapolation of the Orinoco data since the Orinoco watershed is relatively unpopulated compared to much of the region (particularly the Caribbean Islands). Wastewater inputs of nitrogen are estimated as the product of population, the fraction of the population which is sewered (WRI/UNEP 1988), and per capita nitrogen production in wastewater (3.3 kg N yr^{-1} $person^{-1}$; Meybeck et al. 1989). Wastewater inputs of phosphorus were handled similarly, assuming 0.4 kg P yr^{-1} per person (Meybeck et al. 1989), except that the estimate of Ramirez (1991) was used for Venezuela. The percent of the population which is sewered is taken as 70%, 55%, and 50% respectively in the Central American component, the South American component, and the Caribbean islands component of the region (WRI/UNEP 1988).

Amazon and Tocantins

This region includes all of the Amazon River basin plus the basin of the Tocantins River, a river immediately to the south of the Amazon. The region is very large and comprises 36% of South America (Wright 1993). Estimates for area, population, and freshwater discharge are from Meybeck et al. (1989). The Amazon dominates discharge in the region (96% of total discharge) and fluxes of TN and TP. This river is well studied, and very good nutrient flux estimates are available. We used the estimates of Salati et al. (1982) for TN and Devol et al. (1991) for TP. The Amazon data are extrapolated to the Tocantins basin on the basis of discharge. Note that while the Amazon discharges right at the equator, oceanic currents carry its discharge north into the North Atlantic Ocean, so we include it in the North Atlantic basin.

Baltic Sea

The Baltic Sea region includes all drainages flowing into the Baltic (Larsson et al. 1985). Data for area, population, and discharge are from Wulff et al. (1990). Nutrient flux estimates are taken as the total contributions of municipal, industrial and riverine inputs into the sub-basins of the Baltic Sea, excluding nitrogen fixation in the Baltic Sea itself (Larsson et al. 1985).

North Sea

The North Sea region follows the boundaries defined for purposes of nutrient loading by the 1990 North Sea Conference. It includes all Norwegian drainages south of latitude 62° N, all watersheds in the United Kingdom draining into the North Sea and English Channel, and the following watersheds and areas on the continent: Elbe, Weser, Ems, Rhine, Scheldt, Yser, Nord Pas de Calais, Somme, Seine, and Normandy/Brittany. For our overall estimates for the North Sea, we sum separate estimates for population, area, freshwater discharge, and nutrient fluxes for the Norwegian drainages, the U.K. drainages, and the continental drainages. All of our estimates for the Norwegian drainages are from Holtan et al. (1992a). For the U.K. drainages, area is from the Institute of Hydrology/British Geological Survey (1993); population is estimated from this area and the average population density of the U.K. (Espenshade 1990); freshwater discharge is estimated from this area and the average discharge per area of 20 major rivers in the U.K. (Ward 1981). Estimates of regional fluxes of TN and TP for the U.K. portion of this region include river fluxes plus direct sewage and wastewater discharges (HMSO 1992, 1994). For the portion of the North Sea region in continental Europe, population is estimated from national average population densities (Espenshade 1990). Freshwater discharges, and fluxes of TN and TP are taken as the sum of estimates for individual drainages and areas: Elbe, Wester, Ems, Rhine, and Yser Rivers (Rijkswaterstaat 1992); Scheldt estuary (Billen et al. 1985, and unpublished data); Nord Pas de Calais and Somme (Chaussepied et al. 1989; extrapolation from Nord Pas de Calais to Somme based on watershed area); Seine River (Allardi & Billen 1993); Normandy and Brittany (extrapolation for entire area based on watershed areas and data from the Morlaix River; Wafar et al. 1989). Ibrekk et al. (1991) provide independent estimates for TN and TP fluxes to the North Sea region (not including fluxes from France) which are 7% less than ours for TN and 8% greater than ours for TP.

NW European coast

The NW European coast region includes 3 components: Ireland, Norwegian drainages north of Latitude 62° N (including those to the Norwegian and Barents Seas), and the westward drainages of the United Kingdom (including those to the Atlantic, the Irish Sea, and the Celtic Sea). Data and estimates for each of these components are summed to give overall regional estimates. All data and estimates for the Norwegian portion of the region are from Holtan et al. (1992a). Estimates for the U.K. portion of this region are derived as for the U.K. portion in the North Sea, using the same sources. For Ireland, drainage areas and population are national totals (Espenshade 1990), freshwater discharge data are from WRI/UNEP (1988), and fluxes of TN and TP are from the Research Unit of the Department of the Environment of Ireland (ERU 1991).

SW European coast

This region goes from the western boundary of the North Sea through the Guadalquivir drainage of Spain, including watersheds of the following rivers: Loire, Sévre, Charente, Garonne/Dordogne, Adour, Douro, Tagus, Guadiana, and Guadalquivir. Population is estimated by summing data for the individual watersheds using appropriate national average population densities (Espenshade 1990); for drainages that overlap national boundaries, we use an approximate area-weighted average population density. Freshwater discharge and regional fluxes of TN are estimated by summing estimates for the individual river basins: Loire and Sévre (Agence de l'eau Loire-Bretagne 1989); Charente (Zhong Cai (1991); Garonne (Probst 1985); Adour (extrapolated from Garonne based on watershed areas); Tagus and Guadiana (Kempe et al 1991); Douro, Mino, and Guadalquivir (extrapolated from the Tagus and Guadiana using watershed areas and mean areal fluxes). The regional flux for TP is also estimated by summing values for each river basin: Loire and Sévre (Agence de l'eau Loire-Bretagne 1989); Charente, Garonne, and Adour (extrapolated from the Loire and Sévre using watershed areas and mean areal fluxes); Tagus (Kempe et al. 1991); Guadiana, Douro, and Guadalquivir (extrapolated from the Tagus using watershed areas).

NW Africa

The northwestern Africa region includes the drainages from Morocco south to the Zaire River basin, but not including the Zaire (which flows into the South Atlantic). We follow Robertson & Rosswall (1986) and ignore the area of the region in the western Sahara since there is virtually no riverine export there. Drainage area, population, and freshwater discharge are estimated by summing data for the following rivers: Bandama, Cassamance, Cavally, Cestos, Cross, Gambia, Kouilou, Mono, Niger, Ntem, Nyong, Ogooue, St. Paul, Sanaga, Sassandra, Senegal, and Volta (UNESCO 1992). Regional flux of TN is taken from Robertson & Rosswall (1986). The TP flux is estimated from the TN flux by assuming a molar N:P ratio of 38:1 as reported for the Gambia River (Lesack et al. 1984).

Methodological concerns and limitations with river flux estimates

The problems associated with assembling data to construct large-scale estimates of nutrient fluxes have been recognized by previous investigators (Meybeck 1988). We briefly mention a few here to alert the reader to the limitations of our analysis. First, we estimate only annual mean fluxes and ignore inter-annual variability. The nature of such variability for nitrogen fluxes are discussed by Alexander et al. (in press). Second, we have mixed two general approaches for estimating regional freshwater discharge: estimates based on summing riverine flows, and estimates based on regional water balances. The approach of summing individual river flows will tend to underestimate regional discharge by the amount of flow in streams and small rivers which are too small to be reported. Mixing the two estimates of regional flow as we have done is inconsistent, but unavoidable where large data gaps exist. Third, there are obvious errors associated with extrapolation of nutrient fluxes to areas where primary data are not available. We have used a variety of different approaches for extrapolation, such as sometimes using systems which have similar geology, climate, and land use, and other times extrapolating TN fluxes from inorganic nitrogen and organic carbon fluxes. We have used our best judgment in each case, but obviously our use of different approaches introduces some additional inconsistencies. Fourth, where direct estimates of riverine flux of TN and TP are available, they are often based on calculations using monthly data (or sometimes even mean annual values). These values should be interpreted with caution since the concentration of suspended materials is generally correlated with discharge (Muller & Forstner 1968; Lewis & Saunders 1989), leading to underestimates of fluxes calculated with mean values of discharge and concentration. The solution to avoiding biased estimates of nutrient fluxes is to use flow-weighted "average" concentrations, but these are often not available for the systems of interest.

Estimation of anthropogenic nitrogen inputs to regions

We consider four major anthropogenic fluxes into and out of regions: the application of nitrogenous fertilizers, fixation by leguminous crops, the net import or export of nitrogen in agricultural products to or from each region, and atmospheric deposition of nitrogen from pollution sources other than those related to agriculture. Sewage and animal wastes are not considered as inputs to regions as they do not represent newly fixed or imported nitrogen, but rather a redistribution or recycling of nitrogen within a region.

Estimates of fertilizer inputs to basins in Europe, Africa and Latin America are derived from Matthews (1994). The inputs are estimated by taking the reported values per country multiplied by the fraction of that country in each watershed. In some instances, the 1° × 1° map of fertilizer application shown in Matthews (1994) allowed us to determine if all or none of the fertilizer applied to the country occurred within the watershed boundaries, but we did not attempt any finer scaling of per country values. Fertilizer estimates for North American regions are taken from Jordan and Weller (in press), as it is not possible to partition the single U.S. value given in Matthews (1994) among the watershed regions.

We estimate crop nitrogen fixation only for the regions within North America and Europe, partly due to data constraints and partly because these regions are by far the most altered by human activity. Although all types of agricultural land – including pastures – exhibit some amount of nitrogen fixation, we restrict our estimates to five main classes of leguminous crops: i) soybeans, ii) other beans (including broad and dry beans), iii) peas and lentils, iv) peanuts, and v) alfalfa, vetch and lupine. Fixation rates for each of these crop classes were estimated from reviews by Keeney (1979), Stevenson (1982), and Messer & Brezonik (1983). Total fixation inputs for each region are derived by multiplying the rate of fixation for each crop type by the crop area within the watershed. Crop areas for Europe and Canada are taken from 1985 FAO statistics; those for regions within the U.S. come from the 1993 Bureau of Census. As in the fertilizer estimates, the values for Europe are reported on a per country basis, and therefore are scaled for each country by the fraction of country area contained within the watershed boundaries. We do not include nitrogen fixation by non-leguminous crops or pastures largely because of highly variable data on fixation rates (particularly in pastures and rangeland), but also because the range in fixation rates reported for many of these systems (Woodmansee 1978; Messer & Brezonik 1983; Barry et al. 1993) is comparable to that for natural temperate ecosystems (Bormann et al. 1977; Henderson et al. 1978; Boring et al. 1988; Schlesinger 1991). Thus, fixation in such areas is unlikely to be a large source of fixed nitrogen above background natural rates.

Estimates for the net import or export of nitrogen in agricultural products (food and feed) for U.S. portions of the regions are taken from Jordan and Weller (in press). Those for Canada and Europe are estimated by using 1985 FAO statistics on per country imports and exports of the following agricultural categories: i) cereals, ii) leguminous crops, iii) meat, and iv) milk. Average nitrogen contents for each category are taken from Blanck (1955), and per country values are then scaled by watershed areas as above. As for the crop nitrogen fixation estimates, values for net food and feed nitrogen fluxes are derived for only the North American and European watersheds.

The values for deposition of atmospheric nitrogen to each region are described in detail by the atmospheric working group (Prospero et al., this volume). Briefly, Prospero et al. present modeled estimates of wet and dry deposition of oxidized (NO_y) and reduced (NH_x) forms of nitrogen. Those for oxidized nitrogen are based on the GCTM model (Levy & Moxim 1989; Galloway et al. 1994; Levy et al., ms. in prep.), which produces estimates of deposition at a 2.5° × 3.25° latitude-longitude resolution. The NH_x portion is based on a model described by Dentener and Crutzen (in press), which estimates emissions, transport and deposition of ammonia species at 10° × 10° resolution.

Units for presentation

In most of this manuscript, nutrient fluxes are presented in weight units, as is typically done for terrestrial studies. We also provide summaries of overall nutrient inputs to the North Atlantic for each region in molar units to better interface with the oceanographic and coastal working groups of the Block Island workshop. Fluxes per area are given per km^2; for readers more accustomed to hectares, 100 kg km^{-2} = 1 kg $hectare^{-1}$.

Characterization of the regions

Characteristics of the regions bordering the North Atlantic Ocean such as drainage area, population, and freshwater discharge are summarized in Table 1 and Figure 1. Overall, these regions make up 18% of the surface area of all land on earth and have 15% of the world's population. Population density in the regions averages 29 individuals km^{-2}, or 20% less than the world average of 36.5 individuals km^{-2} (Wright 1993). Freshwater discharge from these North Atlantic regions contributes 30 to 33% of the world's total freshwater discharge into the oceans (42 to 46 × 10^{12} m^3 yr^{-1}; Schlesinger 1991; Maidment 1993), with half of this coming from the Amazon (Table 1).

The North Atlantic regions vary greatly in discharge and population density. The Amazon makes up 24% of the total drainage area feeding into the North Atlantic and 50% of the freshwater discharge but has only 1% of the population (Table 1). At the other extreme, the drainages flowing into the North Sea comprise only 3% of the total drainage area and contribute 3% of the freshwater discharge but have 20% of the population. The regions vary in population density from 1.5 person km^{-2} in the Amazon to 186 persons km^{-2} in the North Sea region (Figure 1a). Freshwater discharge per area of watershed varies from 0.12 m^3 m^{-2} yr^{-1} in northwestern Africa to 1.1 m^3 m^{-2} yr^{-1} for northwestern European coast (Figure 1b).

Estimated fluxes of N & P

We estimate nitrogen and phosphorus inputs to the North Atlantic Ocean from terrestrial sources to be approximately 13 Tg N yr^{-1} (935 Gmoles N yr^{-1}) and 2.3 Tg P yr^{-1} (73 Gmoles P yr^{-1}; Table 2). Our estimate does not include inputs into the Mediterranean Sea, which must be quite large: net export of nitrogen from the Mediterranean to the North Atlantic Ocean proper is estimated to be approximately 2.8 Tg N yr^{-1} (200 Gmoles yr^{-1}; Michaels et al., this volume). For nitrogen, our estimate of 13 Tg yr^{-1} is 6.5% of the estimate of the report by the Joint Group of Experts on the Scientific Aspects of Marine Pollution (GESAMP 1987) for riverine input of nitrogen to all the world's oceans, is 32% of total world riverine flux estimate of Galloway et al. (1995), but is 33 to 130% of the total world riverine flux estimated by Smil (1990). The Smil (1990) estimate for the world must clearly be wrong, if one accepts our estimates for the North Atlantic. Our analysis may also suggest that the GESAMP (1987) estimate for the world is high, since the North Atlantic regions have 18% of the world's land surface area, 15% of the world's population, and 30% of the world's freshwater discharge and yet contribute only 6.5% of the GESAMP (1987) estimate for world nitrogen flows into oceans. Nitrogen flux on land (defined as nitrogen use in fertilizer plus livestock and human waste per area) in the North Atlantic regions is typical of that the world as a whole (Nixon 1995). These considerations lead us to favor the estimate of Galloway et al. (1995) as the best currently available for global riverine nitrogen flux to the oceans. Our estimate of P inputs to the North Atlantic from terrestrial sources is 10.5% of the estimated total riverine input of phosphorus to the world's oceans, which is perhaps logical since that global estimate is strongly dominated by the high erosional input of material to the oceans from Asia and Oceania (Howarth et al. 1995).

The Amazon basin dominates the riverine phosphorus inputs to the North Atlantic, making up 68% (Table 2). The Amazon is also the single largest source of nitrogen to the North Atlantic, comprising 25% of the total riverine input (Table 2), although most of the other regions also contribute significantly to nitrogen inputs. In addition to the Amazon, the Mississippi basin, Caribbean and Central American region, the North Sea, and northwestern Africa all contribute more than 1 Tg N yr^{-1} to the North Atlantic (Table 2).

When expressed per area of watershed, nutrient fluxes from the various regions vary markedly (Figure 2a, 2b). The highest fluxes per area for nitrogen are in the watersheds surrounding the North Sea (1,450 kg N km^{-2} yr^{-1}) with high fluxes also along the northwest coast of Europe (1,300 kg N km^{-2} yr^{-1}) and the northeast coast of the United States (1,070 kg N km^{-2} yr^{-1}). The lowest fluxes are from the northern Canadian rivers (76 kg N km^{-2} yr^{-1}). The average nitrogen flux per area for all of the regions flowing into the North Atlantic Ocean is 486 kg N km^{-2} yr^{-1}. Surprisingly, the highest phosphorus flux per area is in the Amazon basin (236 kg P km^{-2} yr^{-1}), followed by the northeast coast of the U.S. (139 kg P km^{-2} yr^{-1}), the watersheds of the North Sea (117 kg P km^{-2} yr^{-1}), and the southwestern European coast (101 kg P

Table 2. Summary of regional data on inputs of N and P to the North Atlantic from land.

	TN		TP		N:P
	Tg yr^{-1}	(Gmol yr^{-1})	Tg yr^{-1}	(Gmol yr^{-1})	(molar)
North Canadian rivers	0.30	(22)	0.018	(0.57)	37
St. Lawrence basin	0.66	(47)	0.02	(0.63)	73
NE coast of US	0.51	(37)	0.067	(2.1)	17
SE coast of US	0.24	(17)	0.011	(0.35)	47
Eastern Gulf of Mexico	0.23	(17)	0.011	(0.35)	46
Mississippi basin	1.82	(130)	0.107	(3.4)	38
Western Gulf of Mexico	0.85	(61)	0.007	(0.22)	271
North America – total	4.61	(331)	0.241	(7.65)	42
Caribbean Islands and Central America (incl. Orinoco)	1.09	(78)	0.141	(4.5)	17
Amazon & Tocantins basin	3.28	(234)	1.53	(49.)	4.8
Central & South America – total	4.37	(312)	1.67	(53.)	5.8
Baltic Sea	0.74	(53)	0.072	(2.3)	23
North Sea	1.22	(87)	0.099	(3.1)	28
NW European coast	0.44	(32)	0.028	(0.89)	36
SW European coast	0.20	(14)	0.054	(1.7)	8.3
Western Europe – Total*	2.60	(186)	0.253	(8.0)	23
NW Africa	1.48	(106)	0.087	(2.8)	38
Total*	13.1	(935)	2.25	(71.)	13

* not including Mediterranean Sea.

km^{-2} yr^{-1}). The lowest phosphorus fluxes per area are from the northern Canadian rivers (4.5 kg P km^{-2} yr^{-1}) and the western Gulf of Mexico region (5 kg P km^{-2} yr^{-1}; Figure 2b). The high phosphorus flux from the Amazon basin is mostly particulate, and is likely a consequence of high rates of erosion and weathering in the geologically young Andean headwaters (Stallard 1985; Devol et al. 1991). Much of this phosphorus flux from the Amazon may consist of apatite, which is insoluble in seawater and therefore considered to be biologically unavailable upon reaching the ocean (Howarth et al. 1995).

N:P ratio of nutrient inputs

The riverine nutrient inputs vary tremendously in their N:P molar ratio among the regions of the North Atlantic basin (Table 2). The highest ratio is found in

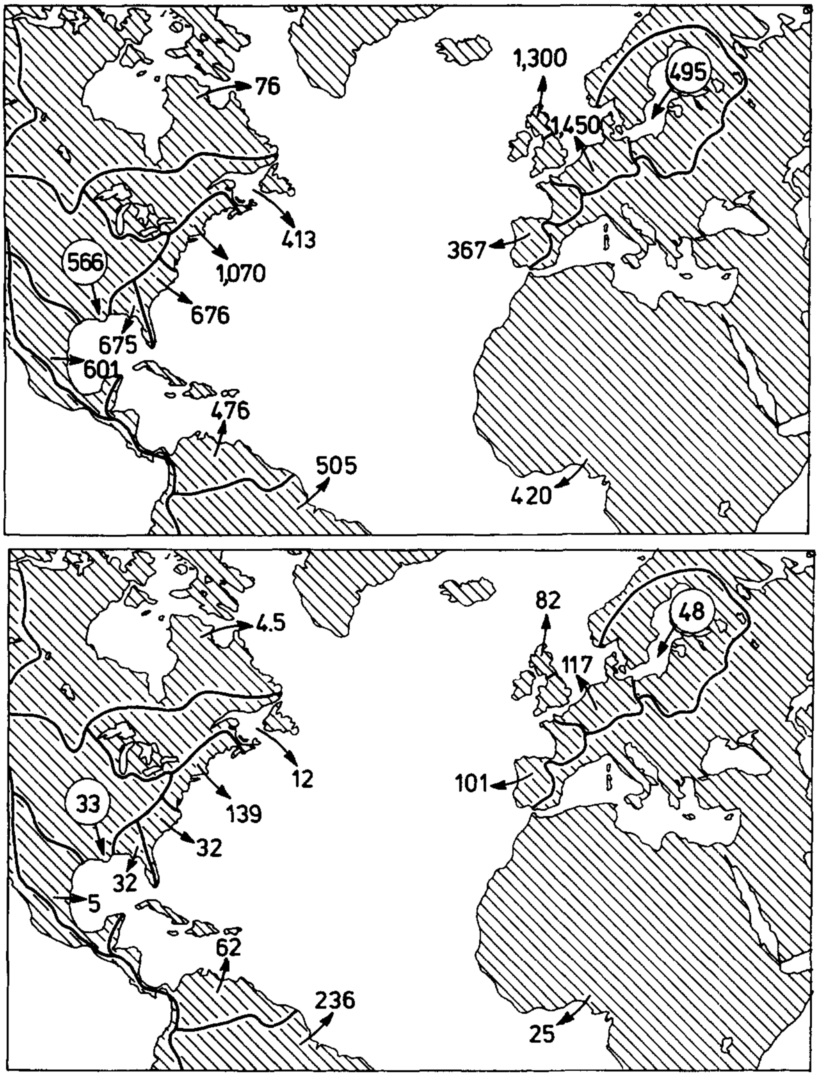

Figure 2. Total nitrogen (TN) export in rivers in kg N km^{-2} yr^{-1} (top, a) and total phosphorus (TP) export in rivers in kg P km^{-2} yr^{-1} (bottom, b). See text for explanation of estimates.

the western Gulf of Mexico (271:1), while the lowest value is in the Amazon basin (4.8:1). The average for the North Atlantic Ocean as a whole (excluding inputs from the Mediterranean Sea) is 13:1 (Table 2). Billen et al. (1991) noted that the N:P ratio of nutrient exports from watersheds is influenced by land use and disturbance, with the ratio for watersheds tending to decrease as total nutrient load increases in response to disturbance. In their analysis, nutrient exports from forested watersheds typically had N:P ratios well above 200:1, exports from agricultural watersheds had ratios between 30:1 and 300:1, and urbanized watersheds had ratios between 5:1 and 40:1. Given the fairly low N:P ratios in Table 2, this analysis might suggest that virtually all of the regions draining into the North Atlantic Ocean are quite disturbed. We find this unlikely, however; note the low population density in the north Canadian rivers region and Amazon basin (Figure 1a), two regions with low to extremely low N:P ratios. We conclude that at the scale of large regions, the N:P ratio of riverine nutrient fluxes is not a good indicator of disturbance. In part, the high N:P ratio noted by Billen et al. (1991) for forested watersheds may reflect high atmospheric deposition onto these forests: two of the three forests they considered are in areas of high atmospheric deposition of nitrogen. As is discussed further below, the nitrogen export (and therefore N:P ratio) from truly undisturbed forests is likely to be much less than observed in forests receiving high rates of nitrogen deposition.

The N:P ratio of nutrient inputs is one important factor influencing whether primary production in estuaries and coastal waters is more likely to be limited by nitrogen or phosphorus (Howarth 1988; Billen et al. 1991; Howarth et al. 1995; Justic et al. 1995). Phytoplankton require nitrogen and phosphorus in the approximate molar ratio of 16:1, the "Redfield" ratio. Consequently, unless there are biogeochemical processes within an ecosystem which alter the N:P ratio (such as nitrogen fixation, denitrification, and phosphorus sorption by sediments), net primary production in ecosystems receiving nutrient loads with an N:P ratio for available nutrients above 16:1 will be phosphorus limited while those receiving loads with an N:P ratio below 16:1 will be nitrogen limited. The N:P ratio for land-based nutrient inputs from most of the regions surrounding the North Atlantic Ocean are within a factor of 2 or 3 of the Redfield ratio. This suggests that estuaries in most of these regions could well be either nitrogen or phosphorus limited, depending upon how biogeochemical processes within the estuary alter the N:P ratio. An important qualification of this argument must be made: not all nutrient fluxes are biologically available, and the N:P ratio for TN and TP fluxes (as reported in this paper) can vary significantly from the N:P ratio for available nitrogen and phosphorus (which is more determining of nutrient limitation). For nitrogen, very little is known about the fate of organic nitrogen inputs to estuaries and coastal oceans; a critical question is to determine how much is biologically available, and how much is simply buried. For phosphorus, Howarth et al. (1995) estimated that only 25% to 70% of the phosphorus inputs to the world's ocean are biogeochemically active, and the rest is biologically unavailable. When human activity increases phosphorus fluxes, the percentage availability of the phosphorus probably also increases (Meybeck 1982; Howarth et al. 1995).

Nutrient fluxes on the continental shelves off most of the North Atlantic regions are dominated by cross-shelf transport from the deep ocean (Michaels et al., this volume; Nixon et al., this volume). Thus, the N:P ratio of riverine nutrient fluxes probably has little influence on whether primary production on much of the shelf area is nitrogen or phosphorus limited. However, riverine inputs can be important in some regions. For example, a large increase in the N:P ratio in discharges from the Mississippi River over the past few decades may have had substantial effects on nutrient limitation on the shelf in the Gulf of Mexico (Justic et al. 1995; Rabalais et al., in press).

Biogeochemical processes within estuaries alter the N:P ratio, with denitrification lowering the ratio and accumulation of phosphorus in sediments raising the ratio. As discussed by the coastal working group (Nixon et al., this volume), rates of denitrification tend to exceed rates of nitrogen fixation in most estuaries; on average, about half of the nitrogen inputs from rivers is consumed within estuaries. Residence time of water within the estuary appears to be the major factor regulating the extent of nitrogen loss through denitrification (Nixon et al., this volume), although there may be a slight trend towards more denitrification in more eutrophic systems. The percentage of phosphorus entering estuaries from rivers that is "permanently" buried in sediments is quite variable, ranging from 10% or less up to more than 100% (in the case of estuaries which are trapping large amounts of phosphorus from offshore; Howarth et al. 1995). Often, the percentage accumulation of phosphorus in sediment decreases as an estuary becomes more eutrophic. Since the percent phosphorus storage is often sensitive to the trophic status of the estuary (with increasing phosphorus availability in more eutrophic estuaries) while nitrogen consumed in an estuary is relatively insensitive to trophic status, biogeochemical processes may be more likely to increase the N:P ratio in pristine oligotrophic estuaries and lower the N:P ratio in more eutrophic estuaries (Howarth et al. 1995). We might expect nitrogen limitation to predominate in many of the North Atlantic regions having high total nutrient fluxes, since estuaries in these regions would tend to be mesotrophic and eutrophic. On the other hand, phosphorus limitation might be more prevalent in the regions having lower total nutrient fluxes, where estuaries are likely to have lower average rates of primary production.

Nutrient limitation in the North Atlantic Ocean as a whole is determined by the total suite of factors influencing the availability of nitrogen and phosphorus. While the riverine input of nutrients has a fairly low N:P ratio (13:1; Table 2), nutrient inputs from the atmosphere tend to have a high N:P ratio, averaging 70:1 for all of the world's oceans (Duce et al. 1991). The importance of nitrogen fixation and losses of nitrogen and phosphorus in the deep ocean may well be the most important factors regulating nitrogen and phosphorus availability (Michaels et al., this volume), with the riverine inputs being quite insignificant to the North Atlantic as a whole.

Direct sources of nitrogen to rivers – sewage and human waste

Human sewage and wastewaters are obvious sources of nitrogen to rivers. In an analysis of 42 major world rivers, Cole et al. (1993) concluded that sewage inputs alone are sufficient to account for the increased flux of nitrate observed in rivers whose watersheds have a higher population density. Although they acknowledged that deforestation, atmospheric deposition, and fertilizer application can all contribute significantly to nitrogen export from rivers, they stated that "watersheds with moderate to high human population will likely be dominated by sewage" rather than other inputs. On the other hand, agricultural sources of nitrogen and nitrogen deposition from the atmosphere are frequently cited as the major causes of increased nitrogen loading to rivers and estuaries (National Research Council 1993). Turner & Rabalais (1991) proposed that the observed increase in nitrogen flux in the Mississippi River over the past few decades is due to increased use of fertilizer in the watershed, and Fisher & Oppenheimer (1991) concluded that atmospheric deposition onto the watershed may be the single largest input to Chesapeake Bay, accounting for 37% of all inputs (Fisher & Oppenheimer 1991).

We estimate the contribution of sewage and wastewater to the exports of nitrogen from each of the regions bordering the North Atlantic Ocean from data on what percentage of the population in each region is sewered. Where available, we use data from OECD (1991). Elsewhere, we use data from WRI/UNEP (1988) on access to waste treatment facilities in urban areas. Actual nitrogen fluxes due to sewage wastewater are calculated from sewered population estimates by assuming a per capita nitrogen load in wastewaters of 3.3 kg N yr^{-1} per person (Meybeck et al. 1989).

The percent contribution of sewage and wastewaters to total nitrogen inputs varies among the regions, from a high of 34% in the North Sea region to virtually none in the Amazon basin (Table 3). The percentage contribution of sewage and wastewaters to total nitrogen flux from a region is strongly correlated with population density ($p < 0.05$; compare Table 3 and Figure 1a). Nonetheless, this analysis suggests that nitrogen sources other than sewage and wastewater inputs dominate fluxes in all regions, and quite strongly in most regions. Overall, nitrogen from sewage and wastewater inputs is only 11% of the total nitrogen inputs to the North Atlantic Ocean from land (Table 3).

Table 3. Sewage and wastewater inputs to the North Atlantic and its river basins.

	Sewage N input ($\mathrm{Tg\ yr^{-1}}$)	% of Total N input
North Canadian rivers	0.022	7%
St. Lawrence basin	0.072	11%
NE coast of US	0.13	26%
SE coast of US	0.034	14%
Eastern Gulf of Mexico	0.049	21%
Mississippi basin	0.16	9%
Western Gulf of Mexico	0.082	10%
North America – total	0.55	12%
Caribbean Islands and Central America (incl. Orinoco)	0.140	13%
Amazon & Tocantins basin	0.0003	0.01%
Central & South America – total	0.14	3%
Baltic Sea	0.10	14%
North Sea	0.42	34%
NW European coast	0.073	16%
SW European coast	0.047	23%
Western Europe – total*	0.64	25%
NW Africa	0.096	6%
Total – all basins*	1.43	11%

* Mediterranean Sea not included.

There may be some error in our assessment of sewered populations, and human waste which does not enter a sewer system can also contribute to nitrogen fluxes from regions to the North Atlantic. To obtain a maximum estimate of human waste contribution to nitrogen inputs to the North Atlantic, we use total population values (Table 1) and an assumption that humans excrete 4.4 kg N $\mathrm{yr^{-1}}$ per person (Vollenweider 1968; note that this is slightly more nitrogen than the per capita load in sewage, referenced above, because of nitrogen losses in sewage treatment). Given the total population in the North Atlantic basin of 790×10^6 (Table 1), the maximum estimate for nitrogen excretion by humans in the regions surrounding the North Atlantic Ocean is 3.5 Tg N $\mathrm{yr^{-1}}$. This is a little over twice our estimate for release of nitrogen in sewage and wastewaters, which seems reasonable since sewage treatment removes some nitrogen and since not all human waste is treated in wastewater treatment plants. Even if all human waste reached the North Atlantic, it could account for at most 27% of total nitrogen inputs (Tables 2 and 3). For the North Sea region, the most populous region surrounding the North Atlantic, we similarly estimate that total nitrogen production in human waste might be 0.69 Tg N yr^{-1}, or roughly half of the total regional flux of nitrogen to the ocean (Tables 2 and 3). Clearly non-sewage sources of nitrogen dominate fluxes from all regions. These non-point sources include agricultural runoff, atmospheric deposition onto watersheds, and runoff from animal feedlots.

Peierls et al. (1991) noted that on a log-log plot, nitrate fluxes in the major rivers of the world are correlated with human population density (Figure 3). Total nitrogen fluxes per area for the 14 regions of the North Atlantic basin are also correlated with human population density on a log-log scale ($r^2 = 0.45$; $p = 0.01$; Figure 3). However, the slope of the relationship for total nitrogen fluxes (our data for North Atlantic regions) is less than that for nitrate fluxes in major world rivers (Peierls et al. 1991) by almost a factor of 2 (Figure 3). This suggests that disturbance associated with human population density preferentially mobilizes nitrate over other nitrogen forms. These correlations between population density and either nitrate or total nitrogen fluxes could reflect the importance of human waste as one nitrogen input to watersheds or could reflect correlations between human population density and accelerated nitrogen cycling through fertilizer use, food movements, atmospheric pollution, and land disturbance. This latter explanation is perhaps more likely since non-point sources of nitrogen dominate fluxes of nitrogen in the rivers of the North Atlantic basin.

Net nitrogen inputs to the regions from human activity

We estimate total net anthropogenic nitrogen inputs to the watersheds of the North Atlantic basin to be approximately 30 Tg N yr^{-1} (calculated from input date in Table 4 and areas in Table 1); this value includes leguminous crop fixation and net fluxes of food and feed for temperate but not for tropical regions. This represents an extremely large flux of nitrogen into these watersheds; it is 2.5 fold greater than the total river flux to the North Atlantic (Table 2), and roughly one-third of global nitrogen fixation from all natural sources (Soderlund & Rosswall 1982). Fertilizer and deposition dominate total inputs, accounting for 21.8 and 8.2 Tg N yr^{-1}, respectively. Fixation by leguminous crops (4.6 Tg N yr^{-1}) and net food and feed movement (-3.8 Tg N yr^{-1}) nearly balance at this scale.

For atmospheric inputs of fixed nitrogen to each region from human activity, we consider only deposition of oxidized nitrogen compounds (NO_y, including both wet and dry deposition), and only the increase in this deposition

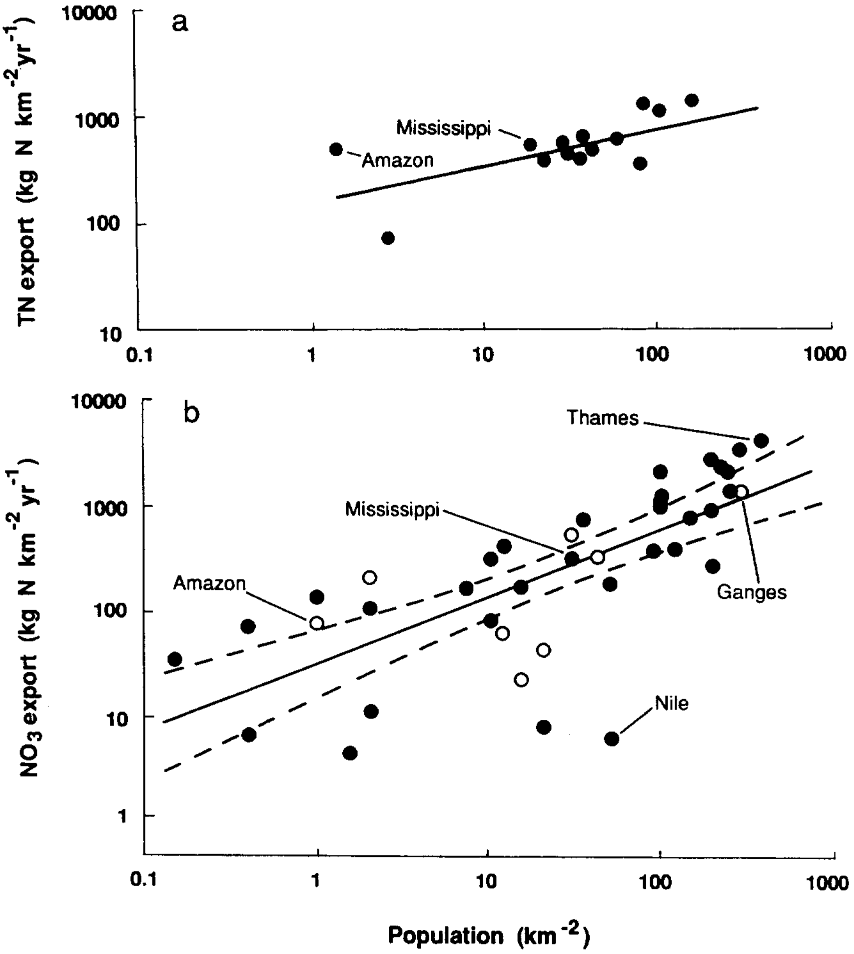

Figure 3. Relationship between log of population density and log of total nitrogen export from regions of the North Atlantic basin (top, a) and of nitrate in major world rivers (bottom, b; redrawn and reprinted by permission from Peierls et al. 1991). Both relationships are significant, but the relationship for nitrate fluxes in world's rivers (bottom) is more significant, has less scatter, and has a steeper slope. For TN fluxes in the North Atlantic basin (top), log TN = 2.2 + 0.35 log (population density); $r^2 = 0.45$; $p = 0.01$. For nitrate fluxes in world rivers (bottom), log NO_3^- = 1.15 + 0.62 log (population density); $r^2 = 0.53$; $p = 0.00001$.

due to human activity; we estimated this by subtracting modeled estimates of pre-industrial deposition from those for current levels estimated by Prospero et al. (this volume). Estimates for modern and pre-industrial deposition of both NO_y and NH_x are shown in Table 5. NH_x deposition is excluded from the estimate of atmospheric nitrogen input to a region because ammonia and ammonium do not travel far in the atmosphere before being deposited back to the ground, and because the principal sources of high levels of ammonia in the atmosphere are volatilization from fertilizer and animal waste products (Schlesinger & Hartley 1991). Both of these sources are accounted for as inputs to a region in our estimates of fertilizer, nitrogen fixation by crops, and net movement of nitrogen in feeds. Adding NH_x deposition would be a form of double accounting and would cause an over-estimate of total nitrogen inputs at the scale of regions since most of the NH_x deposition is driven by NH_x volatilization in the same region.

In contrast to NH_x, the principal sources of NO_y are probably from combustion of fossil fuels and are not otherwise accounted for as inputs of nitrogen to a region. Some NO_y may come from soil processes, such as denitrification, but we suspect that this is small compared to the fossil-fuel source. That NO_y deposition is largely independent of our other source estimates and is an additional input to each region, while NH_x deposition is related to other inputs, can be seen by regressing both NO_y and NH_x deposition against our estimates for fertilizer inputs to a region: NO_y deposition shows no relationship, whereas at the scale of these large regions, NH_x deposition and fertilizer inputs are significantly correlated ($r^2 = 0.53$; $p < 0.05$; data in Table 5 and Figure 2a). It is important to note, however, that while at the scale of an entire region NH_x deposition is not an additional new input of nitrogen, volatilization and subsequent deposition of agricultural nitrogen can cause significant redistribution of nitrogen within a region, much of which may fall on forests and other natural ecosystems and be transported into rivers. This is discussed below in the section on "Fates of Anthropogenic N in Non-Agricultural Lands."

As with the river nitrogen fluxes, there is substantial variation among the regions in the intensity of nitrogen inputs. Human alteration of the nitrogen cycle is clearly much greater in the temperate portions of the North Atlantic basin, with nearly 90% of total anthropogenic nitrogen inputs occurring in the temperate-zone North American and European watersheds (Table 4). The enormous range among the watersheds in the intensity of human disturbance becomes especially clear when inputs are expressed on a per area basis: in the Amazon basin and the Central America/Caribbean region, total net anthropogenic nitrogen inputs average 174 and 482 kg N km^{-2} yr^{-1}, while in North America they average 1,389 kg N km^{-2} yr^{-1} (Table 4). From this perspective, the European regions are the most altered; their average total net anthropogenic input of 3,913 kg N km^{-2} yr^{-1} is more than twice that of the North American watersheds and an order of magnitude greater than the tropical regions (Table 4). There is a tremendous range in total inputs among individual watershed regions even within the temperate zone, from a low in Northern Canada of 216 kg N km^{-2} yr^{-1} to a high of 7,044 kg N km^{-2} yr^{-1} in the North Sea (Table 4).

Table 4. Major net anthropogenic nitrogen inputs to the North Atlantic watersheds by region. All values are in kg N km^{-2} yr^{-1}.

	Anthropogenic NO_y deposition	N fertilizer	Leguminous crop fixation	Import (+) or Export (−) of food and feeds	Total inputs
North Canadian rivers	72.	161.	32.9	−50.2	216.
St. Lawrence basin	612.	331.	256.	−31.3	1,168.
NE coast of US	1,204.	600.	748.	998.	3,550.
SE coast of US	1,023.	1,170.	369.	454.	3,016.
Eastern Gulf of Mexico	763.	1,260.	248.	576.	2,847.
Mississippi basin	620.	1,840.	1,060.	−1,300	2,220.
Western Gulf of Mexico	318.	1,254.			1,572.
North America – total	431.	878.	397.	−317.	1,389.
Caribbean Islands and Central America	140.	342.			482.
Amazon and Tocantins basin	111.	63.			174.
Central and South America – total		118.	136.		254.
Baltic Sea	479.	1,730.	26.7	20.7	2,256.
North Sea	1,085.	5,960.	4.7	−4.2	7,044.
NW European Coast	1,087.	2,870.	55.0	−324.	3,688.
SW European Coast	462.	3,370.	14.7	−64.0	3,783.
Western Europe – total	698.	3,230.	21.9	−36.5	3,913.

Note: Total N inputs for the Western Gulf region and all of Latin America do not include estimates of crop fixation or of net food and feed movement; thus, the continental totals are based on somewhat different components for each continent.

Table 5. Atmospheric deposition of oxidized (NO_y) and reduced (NH_x) forms of nitrogen to the watershed regions. Modern values are total current deposition as reported in Prospero et al. (this volume), pre-industrial values are modeled estimates described in text, and anthropogenic values are the difference between modern and pre-industrial. All values are in kgN $km^{-2}\ yr^{-1}$.

	Modern NO_y	Pre-industrial NO_y	Anthropogenic NO_y	Modern NH_x	Pre-industrial NH_x	Anthropogenic NH_x
North Canadian rivers	84.0	11.6	72.4	70.0	28.7	41.3
St. Lawrence basin	630.	18.3	612.	266.	43.3	223.
NE coast of US	1,232.	28.0	1,204.	238.	43.7	194.
SE coast of US	1,078.	54.6	1,023.	350.	55.5	295.
Eastern Gulf of Mexico	812.	49.0	763.	280.	58.5	222.
Mississippi basin	658.	37.8	620.	266.	45.8	220.
Western Gulf of Mexico	364.	46.2	318.	224.	28.7	195.
Caribbean Islands and Central America	210.	70.0	140.	182.	83.2	98.8
Amazon and Tocantins basin	196.	85.4	111.	140.	86.7	53.3
Baltic Sea	490.	11.3	479.	560.	47.5	513.
North Sea	1,092.	7.3	1,085.	742.	57.5	684.
NW European coast	1,092.	4.8	1,087.	742.	60.1	682.
SW European coast	476.	14.0	462.	322.	53.9	268.

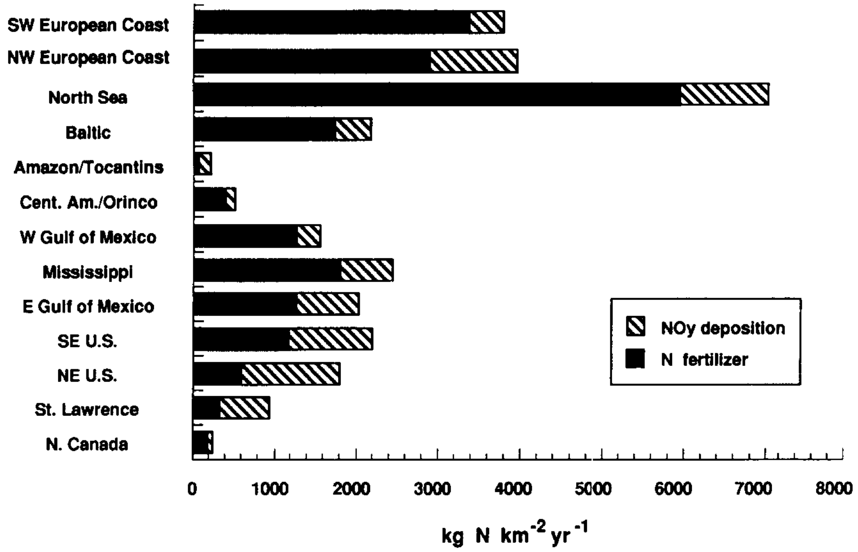

Figure 4. Application of nitrogen in fertilizers and by atmospheric deposition of NO_y to 13 of the watershed regions. Values are in kg N km^{-2} yr^{-1}. Fertilizer inputs dominate net anthropogenic nitrogen inputs in most, but not all, regions of the North Atlantic basin.

The relative importance of different anthropogenic nitrogen sources also varies substantially among the individual watersheds. Nitrogen fertilizer is the major input to most regions, although inputs through atmospheric deposition of NO_y are greater in the St. Lawrence basin and the northeast and southeast coastal regions of the United States (Table 4). The net movement of nitrogen in agricultural food stocks is important only in the northeastern United States, where it is a major input, and the Mississippi basin, where it is a major export. While never the major input, nitrogen fixation by leguminous crops is often a significant input in the North American regions but is generally quite minor in the European regions (Table 4; Isermann 1993). That agricultural sources are dominant inputs to the Mississippi basin is expected since this region drains the heartland of American agriculture. More surprising is the fertilizer dominance in Europe, despite notoriously high rates of atmospheric deposition in some areas; fertilizer inputs dominate in all of the European regions, accounting for 83% of total net anthropogenic inputs to Europe as a whole. Much of the atmospheric deposition in Europe is deposition of NH_x which originates from fertilizer and animal wastes (and we consider to be recycled nitrogen within the region). Per area rates of fertilizer application are far higher in Europe than elsewhere (Figure 4; Table 4).

Comparison of riverine fluxes with anthropogenic inputs – temperate regions

This section focuses on a comparison of net anthropogenic inputs of nitrogen and river fluxes for regions in the temperate regions only. The tropical regions within the North Atlantic basin, in our opinion, are currently much less disturbed and are likely to respond to changing inputs much differently than temperate ecosystems. Further, natural fluxes appear to be greater in the tropics. Fluxes in the tropics are discussed separately below in the section on "Nitrogen Balance in the Humid Tropics."

Fluxes of total nitrogen per area from the temperate regions of the North Atlantic basin are strongly correlated with net anthropogenic inputs of nitrogen per area to these regions ($r^2 = 0.73$; $p = 0.002$; Figure 5a). Note that Figure 5a shows the relationship between linear river fluxes and linear inputs, rather than the weaker log-log relationship between river fluxes of nitrogen and human population density shown in Figure 3. To analyze which nitrogen inputs are most related to riverine fluxes, we can separate the total anthropogenic fluxes into two categories: those related to the combustion of fossil fuels (represented by NO_y deposition) and agricultural inputs (consisting of fertilizer inputs, nitrogen fixation by crops, and the net movement of nitrogen in feeds). The resultant two variable linear model (see Figure 5b) shows that both the sum of the agricultural inputs and NO_y deposition are significant predictors of river nitrogen export ($R^2 = 0.89$, $p = 0.0005$). Given that NO_y deposition is a much smaller input than agricultural sources in most regions, and that nitrogen limitation of production is prevalent in the natural systems throughout most of these regions, we find it especially intriguing that deposition is so well correlated with river fluxes. This point is further illustrated in Figures 5c and 5d, which compare simple regressions of river nitrogen export against NO_y deposition ($r^2 = 0.79$; $p = 0.0006$) and fertilizer ($r^2 = 0.39$; $p = 0.05$), respectively. While neither leguminous fixation nor the net movement of nitrogen in feeds was a significant individual variable in any of the analyses, it is clear from the regional data that they may be important in some watersheds, such as the Mississippi (see Table 4).

The strength of these correlations seems somewhat surprising, considering the wide range in magnitude of fluxes and inherent limitations with the quality of data. Given the enormous range in nitrogen inputs being considered, it is even more surprising that the correlations suggest a *linear* relationship between net anthropogenic inputs and riverine fluxes. The capacity for processes within the landscape to provide a "sink" for excess nitrogen before it reaches the coasts seems inherently limited; therefore, one might expect to see river nitrogen fluxes increase exponentially with increasing nitrogen inputs,

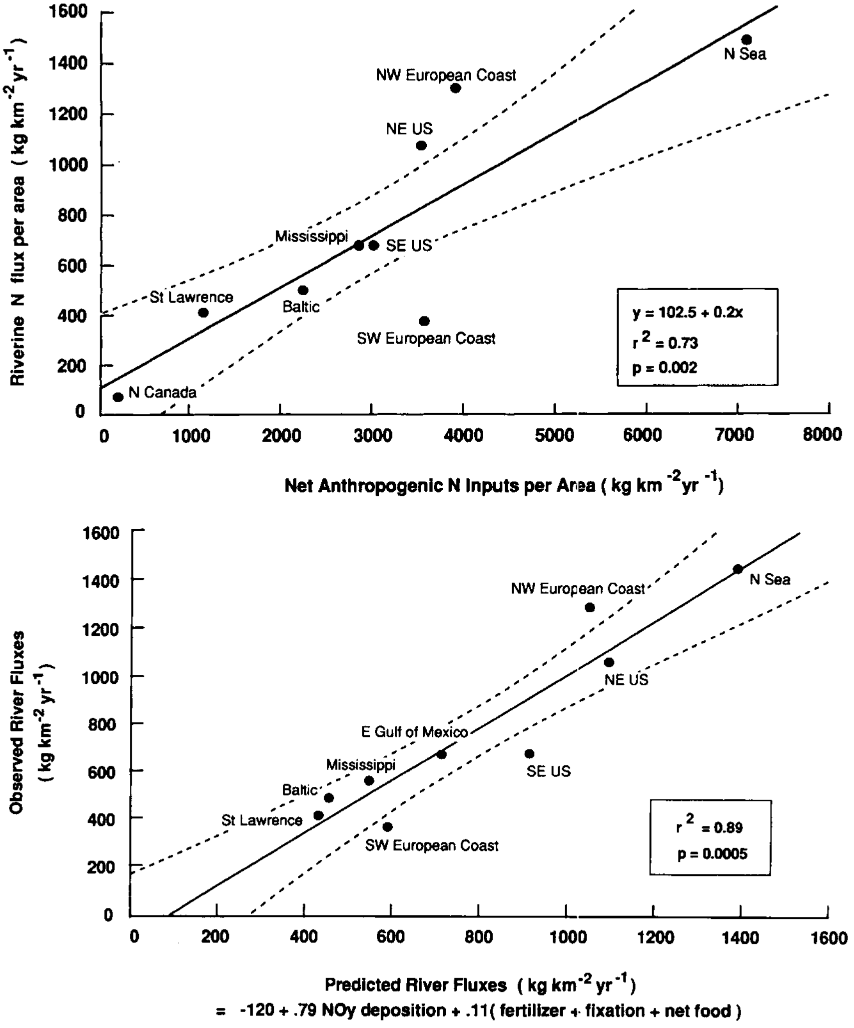

Figure 5. A. Plot of river nitrogen export versus net human-derived nitrogen inputs to each of the temperate regions (all European and North American regions, except the Western Gulf of Mexico); net inputs are equal to the sum of anthropogenic NO_y deposition, fertilizer inputs, nitrogen fixation by crops, and the net import or export of nitrogen in food and feed.

B. Modeled versus observed river nitrogen export for the same set of regions as in 5A. The model is a multiple linear regression of river nitrogen flux against "agricultural" inputs (fertilizer, plus nitrogen fixation by leguminous crops, plus net import or export of nitrogen in food and feed) and inputs from combustion of fossil fuels (anthropogenic NO_y deposition).

yet we find highly significant linear relationships across a 30-fold range in per area inputs.

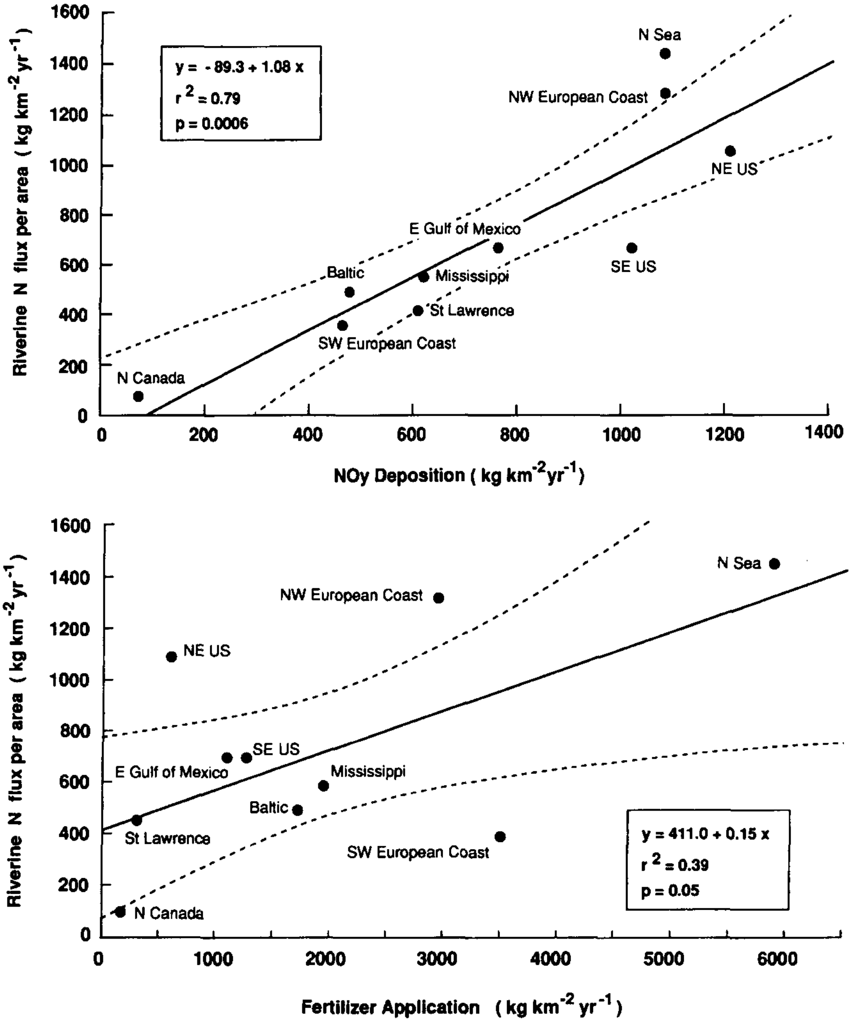

Figure 5. Continued.

C. Simple regression of river nitrogen export per area versus anthropogenic NO_y deposition for the same regions as in 5A.

D. Same as for 5C, but regressor is application of nitrogen fertilizer to each region.

The ratio of riverine nitrogen flux to net anthropogenic nitrogen inputs to the temperate regions of the North Atlantic basin is consistently low, ranging from 0.1 (southwestern European coast) to 0.35 (northwestern European coast, St. Lawrence basin, and northern Canadian rivers region) and averages 0.25 (calculated from Tables 2 and 4). That is, on average 75% of the net anthropogenic nitrogen inputs do not appear in riverine fluxes but rather must be denitrified or stored in the landscape; the total amount of nitrogen removed or stored in the landscape is even greater than this since some nitrogen is also input to each region through natural processes. Careful mass balance studies of partially forested watersheds with substantial agricultural activity have reached similar conclusions, finding some 80% of net anthropogenic nitrogen inputs stored within the watershed or denitrified and only 20% exported in rivers (Jaworski et al. 1992; Lowrance & Leonard 1988).

It is critical to identify the processes behind the apparent pattern of anthropogenic nitrogen loss upstream from the coastal estuaries, as some pathways may be relatively stable across a large range of inputs whereas others may quickly saturate. Denitrification loss as N_2 is the most benign and perhaps the least limited pathway; Neff et al. (1994) present evidence of increasing denitrification in an upland terrestrial system with increases in nitrogen deposition. However, changes in the landscape such as removal of wetland and riparian areas could reduce the total denitrification capacity of a given region (Krug 1993; Downing et al. 1993). Other nitrogen sinks include storage in groundwater and storage in organic matter in terrestrial ecosystems. Both of these might tend to saturate with time and continued high rates of nitrogen input, perhaps eventually resulting in higher rates of riverine nitrogen flux to the North Atlantic.

Fate of fertilizer N in agricultural lands

Fertilizer is the single largest input of nitrogen to the watersheds of the North Atlantic basin, and so it is important to determine the fate of nitrogen applied in fertilizer. In general, somewhat less than 50% of nitrogen fertilizer applied to fields is removed in crop harvest, and the rest is stored in the soil, is volatilized to the atmosphere, or moves into groundwater or surface waters (Bock 1984; Nelson 1985). Obviously, movement of nitrogen into surface waters is one pathway for increasing riverine fluxes, and the magnitude of this pathway is discussed below. However, nitrogen which is harvested in crops can also affect riverine fluxes at the scale of the region, as crops are fed to livestock or to humans with resulting nitrogen flows in wastes. Nitrogen volatilized into the atmosphere from agriculture lands can also affect riverine fluxes as it is deposited back onto terrestrial systems elsewhere in the region. This and the storage of nitrogen in groundwater are discussed in later sections.

A variety of factors can influence the rate of leaching of nitrogen from agricultural systems into surface waters, including water runoff, rate of fertilizer application, type and texture of soil, and the type of land use. Figure 6 summarizes a number of small watershed or lysimeter studies aimed at quantifying nitrogen losses to the hydrosystem from agricultural systems within the North

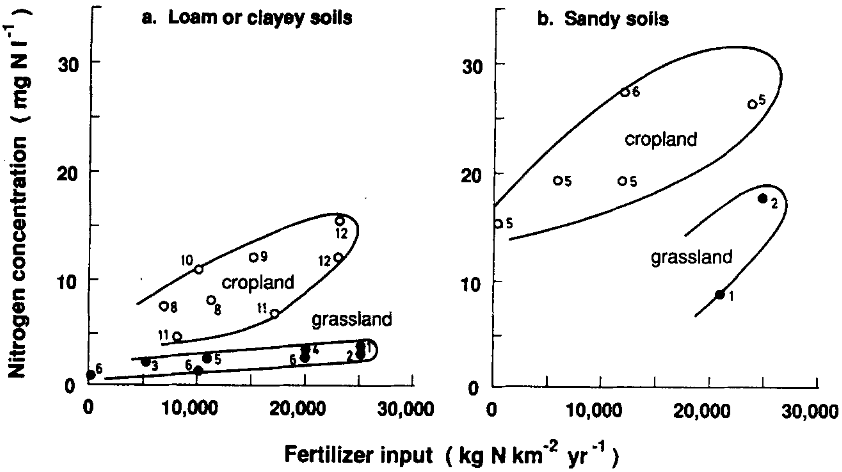

Figure 6. Mean total nitrogen concentration in water leached from agricultural watersheds of different soil type and varying rate of nitrogen fertilizer application. a) Cropland and grassland over loamy or clayey soils. b) Cropland and grassland over sandy soils.

a. Grassland and cropland over loamy and clayey soils: 1. Meadow (NL) (Jurgens-Gschwind & Jung 1979); 2. Meadow (NL) (Kolenbrander 1969); 3. Meadow (B) (De Becker et al. 1984); 4. Grassland (UK) (Webb & Walling 1985); 5. Grassland (UK) (Smith et al. 1982); 6. Meadow (Cz) (Ulehlova 1987); 7. Cropland (NL) (Kolenbrander 1973); 8. Cropland (B) (De Becker et al. 1984); 9. Cereals (F) (Mariotti 1982); 10. Cereals (F) (Belamie 1980); 11. Cereals (Cz) (Bashkin 1987); 12. Corn (England) (Gold et al. 1990); 13. Bluegrass (Ontario) (Baldwin et al., 1977); 14. Corn (Ontario) (Baldwin et al. 1977); 15. Corn (Vermont) (Baldwin et al.,1977); 16. Corn (Ohio) (Baldwin et al. 1977).

b. Grassland and cropland over sandy soil: 1. Meadows (B) (De Becker et al. 1984); 2. Meadows (NL) (Kolenbrander 1973) 3. Arable land (NL) (Sluijsmans et al. 1978); 4. Coastal farmland (S) (Fleisher et al. 1987); 5. Cereals (Sweden) (Bertilsson 1988); 6. Cereals (G) (Strebel et al. 1984); 7. Corn/oats (Iowa) (Baldwin et al. 1977).

Atlantic watershed. Runoff-weighted average concentration of total nitrogen in leachate is plotted against fertilizer inputs. Collectively, these data clearly illustrate the much higher susceptibility to nitrogen leaching of sandy versus loamy or clayey soils, and of arable land compared to grasslands. For runoff rates between 0.2 and 0.7 m^{-3} m^{-2} yr^{-1}, and for loamy or clayey soils, nitrogen export varies between 3 and 10% of fertilizer input for grasslands, and between 10 and 40% for arable land. On sandy soils, corresponding figures are 15 to 50% for grasslands and 25 to 80% for arable land. These percentages are misleading, however, because other sources of nitrogen than added fertilizers (such as atmospheric deposition and nitrogen fixation) contribute to the nitrogen economy of agricultural lands and because leaching losses are not necessarily proportional to total nitrogen input. Relatively high leaching rates (compared to those observed for forests or unfertilized grassland) are often observed for arable land receiving no fertilizer input, perhaps due to lowered evapotranspiration. There exists evidence that this was already the case in agricultural systems even before the advent of inorganic fertilizer use. Thus, a long-term set of observations of the water composition of the Seine River upstream from Paris (Naves et al. 1991; Meybeck 1993) shows that by the beginning of the 20th century (when fertilizer input and atmospheric deposition were negligible), nitrate concentrations were already about 100 μM, or about 5 times higher than the concentration presently found in forested watersheds in the same region (Thibert 1994), probably exposed to higher atmospheric inputs of nitrogen than in 1900.

Using the data in Figure 6, as well as the mean runoff rate, agricultural land use, fertilizer application and soil types of the different North Atlantic watershed regions, we can roughly estimate the potential magnitude of leaching of fertilizer nitrogen from agricultural lands in temperate regions. We are unable to make such a calculation for tropical regions because of a lack of data on soil leaching rates for agricultural systems in these areas. For Europe, we estimate that nitrogen leaching from agricultural soils is approximately 3.6 Tg N yr^{-1} (Table 6), or nearly one third of the total net nitrogen inputs to the region (Table 4) and 138% of our estimated total delivery to the coastal zone in rivers (Table 2), suggesting that some further removal processes occur within the river system (see below). For North America, our estimated leaching rate of agricultural soil is 2.2 Tg N yr^{-1} (Table 6), or only half the total riverine delivery of 4.6 Tg N yr^{-1} (Table 2), stressing the importance of sources of nitrogen other than leaching of fertilizer from agricultural lands. These other sources might include nitrogen applied in fertilizer, but then harvested as crops or volatilized to the atmosphere, as well as nitrogen from fossil-fuel combustion.

Fates of anthropogenic N in non-agricultural lands

Anthropogenic nitrogen reaches non-agricultural lands largely by deposition from the atmosphere, including both NO_y and NH_x (which although not considered here to be a net anthropogenic input to a region can be a significant internal flux of nitrogen to non-agricultural lands; Table 5). Forests and other non-agricultural lands have the potential to store nitrogen in vegetation and soils. Prior to human influence, most of the forest and grassland systems in our study region were probably nitrogen limited (Vitousek & Howarth 1991); thus, some amount of nitrogen inputs from the atmosphere might be tightly retained in vegetation and soils. However, as such systems are continuously exposed to elevated levels of deposition, their capacity for retention of those inputs may "saturate," and losses of nitrogen in both aqueous and gaseous

Table 6. Estimated inputs of nitrogen to surface water by leaching from agricultural soils in the temperate watershed regions of the North Atlantic basin.

	Percent crop land	Percent grass land	Nitrogen concentration in cropland runoff (μM)	Nitrogen concentration in grass land	Runoff $m^3\ m^{-2}\ yr^{-1}$	Nitrogen export $Tg\ N\ yr^{-1}$
St. Lawrence basin	20%	3%	290	70	500	0.66
NE coast of US	11%	4%	430	110	430	0.15
SE coast of US	13%	6%	500	140	170	0.06
Eastern Gulf of Mexico	9%	11%	500	140	300	0.09
Mississippi basin	32%	30%	360	70	170	1.04
Western Gulf of Mexico	8%	44%	360	70	200	0.24
North America	14%	15%			290	2.24
Baltic Sea	9%	1%	1,800	710	320	1.11
North Sea	33%	29%	710	140	450	1.45
NW European coast	6%	10%	1,300	710	1,110	0.76
SW European coast	40%	25%	360	140	200	0.26
Europe	20%	13%			420	3.58

Note: assumed mean concentration in leachate deduced from the data of figure 6, according to the mean fertilizer application rate and the dominant type of soil; Baltic soils are sandy, SW European soils are loamy or clayey, North Sea is one-third sandy and two-thirds loamy or clayey, and NW Europe is two-thirds sandy and one-third clayey or loamy.

forms may increase rapidly (Aber et al. 1989; Gundersen & Bashkin 1994). A number of studies have shown extremely high nitrogen leaching losses from European forests in high deposition regions (van Breemen et al. 1982; Schulze et al. 1989; Durka et al. 1994). A switch from nitrogen retention and accumulation to high nitrogen losses may also be a self-accelerating process: rising concentrations of mineral nitrogen can lead to soil acidification and parallel increases in concentrations of potentially toxic cations such as aluminum, possibly leading to ecosystem decline and even greater nitrogen losses (Skibe et al. 1989; Schulze et al. 1989; Willison et al. 1990). In addition, high nitrogen deposition is often associated with diminishing availability of other elements such as magnesium or calcium. This may limit the ability of vegetation to take up nitrogen and thereby further accelerate nitrogen losses and system decline (Schulze 1989), although there is no evidence for increasing concentrations of base cations in rivers of the northeastern U.S. (Jaworski and Howarth, ms. in prep.).

At the scale of large regions, it is very difficult to predict what fraction of nitrogen deposition onto the natural portion of the terrestrial landscape might reach the aquatic systems. In a review of European data, Hauhs et al. (1989) showed that leaching losses of nitrogen from forests tended to be either very small (<100 kg N km^{-2} yr^{-1}) or relatively large (>500 kg N km^{-2} yr^{-1}). Johnson (1992) summarized data on a number of U.S. forests in which nitrogen losses were shown to vary from a tiny fraction to more than 100% of total inputs, with no apparent relationship to the size of the inputs. However, Hedin et al. (1995) point out that many attempts to use stream export of nitrogen as an index of forest nitrogen retention have not measured organic or particulate nitrogen. In their Chilean sites, organic nitrogen is the major form of export. Thus, the lack of a correlation between inputs and losses in Johnson (1992) may in part arise from a mix of studies in which not all forms of stream nitrogen were measured. In addition, some of the variability may be due to the fact that retention in any given system may depend not only on the amount and prior history of deposition, but also on species composition, soil type, and successional stage (Vitousek & Reiners 1975). Young forests which are accumulating biomass may retain much of the available nitrogen even in areas of high inputs, whereas more mature stands can show losses of nitrogen equal to or greater than inputs even in areas of low deposition (Johnson 1992). However, even young forests can show high nitrogen losses; Lajtha et al. (1995) showed that only 40 to 62% of nitrogen inputs were retained in a young, N-limited forest on Cape Cod, Massachusetts, USA. Losses from neighboring, mature forests were similar, and the authors concluded that the sandy soils of these forests "may have an inherently low ability to retain nitrogen" at any successional stage. The seasonal timing of inputs may also

Table 7. Estimate of maximum possible retention by forests of net anthropogenic nitrogen additions in temperate-zone regions.

	Percent land forested	Increased deposition due to human activity (NO_y and NH_x) (kg N km^{-2} yr^{-1})	Percent contribution of anthropogenic NO_y and NH_x deposition to total net anthropogenic N inputs	Maximum percent of net anthropogenic N inputs to regions retained in forests
North Canadian rivers		114.	53%	
St. Lawrence basin	77%	835.	71%	55%
NE coast of US	85%	1,398.	39%	33%
SE coast of US	81%	1,318.	44%	36%
Eastern Gulfof Mexico	80%	985.	35%	28%
Mississippi basin	38%	840.	38%	14%
Western Gulf of Mexico	48%	513.	33%	16%
Baltic Sea	90%	992.	44%	40%
North Sea	38%	1,769.	25%	10%
NW European coast	84%	1,769.	48%	40%
SW European coast	35%	730.	19%	7%
Average*	70%	730.	37%	26%

* Assuming 90% of northern Canadian region is forested.

influence nitrogen losses; in the Cape Cod sites uptake and retention were high in the summer months, but losses tended to equal or even exceed inputs during the dormant season.

The nitrogen added to a region through human activity can be stored in forests only to the extent that it is deposited on forests. By assuming that all nitrogen deposited onto a forest is retained within a forest, we can estimate the maximum percentage of "anthropogenic nitrogen" which may be thus stored. In Table 7 we show such estimates for each of the regions of the North Atlantic basin that are in the temperate zone. Note that for these calculations, deposition of both NO_y and NH_x above background pristine levels are used; including the NH_x deposition accounts for the volatilization of ammonia from agricultural soils and feedlots within a region with subsequent deposition onto forests. This is recycling within the region, and not a new input of nitrogen to the region, but is critical in determining whether anthropogenically added nitrogen may be stored in forests.

The amount of the net anthropogenic nitrogen added to each region which is deposited from the atmosphere (including the redistribution of NH_x) varies from a low of 19% in southwestern Europe to 71% in the St. Lawrence watershed. By multiplying these values by the percent land area in each region which is forested, and assuming that deposition on forested land occurs at the same rate as on non-forested land, we can estimate the percentage of the net anthropogenic nitrogen added to each region which is deposited onto forests (above background levels of deposition). This is the maximum amount of the net anthropogenic nitrogen which could potentially be stored in forests, assuming that no nitrogen leaves forests. This percentage varies from only 7% in southwestern Europe to 55% in the St. Lawrence watershed (Table 7). From similar calculations for the entire land mass in the temperate zone of the North Atlantic basin, we conclude that a maximum of 26% of net anthropogenic nitrogen might be retained and stored in forests. While not trivial, this is only one third of the amount of net anthropogenic nitrogen which is not exported to the North Atlantic in rivers (see discussion in "Comparison of Riverine Fluxes with Anthropogenic Inputs – Temperate Regions," above). Clearly, the nitrogen added to the North Atlantic basin through human activity is not only stored in forests or exported in rivers; a large amount of nitrogen which never contacts forests must also be stored or denitrified elsewhere in the landscape.

Storage of nitrogen in groundwater

In assessing the fate of increased nitrogen inputs to the terrestrial systems of the North Atlantic drainage basin, the increased storage of nitrogen in groundwater must be considered. A rapid increase in nitrate concentrations

Table 8. Rate of increase of nitrate concentration in groundwaters during the 1980's in different areas in Northwestern Europe and North America. Some data from rivers during periods of baseflow are also included; these are marked by *.

	Aquifer Characteristic	Nitrate increase (μM yr^{-1})	Reference
Europe			
Jutland (DK)	Sand	6.2	Overgaard 1984
Limburg (NL)	Sand	36.	Strebel et al. 1989
Bayern (G)	Sand/gravel	24.	Resch 1991
Wallongy (B)	Sand	26.	Fond. Roi Baudouin 1992
"	Chalk	24.	"
"	Limestone	10.	"
Champagne	Chalk	34.	Strebel et al. 1989
Seine River basin*		5.7	Naves et al. 1991
North America			
Mississippi River basin*		5.0	Turner and Rabalais 1991
South Wisconsin	Sandstone	7.9	Mason et al. 1990
Nebraska	Alluvial	36.	Schepers et al. 1983

in groundwater has become a major concern in most areas with intensive agriculture in both Europe and North America (Table 8). Schroder (1985) observed that the rate of increase in nitrate contamination of groundwater in Denmark is approximately the same as found in the streams. Although this might not be true in all situations, some data on the rate of increase in river nitrate concentration, measured at base flow, are included in Table 8 for areas where groundwater data were not available. The data presented by Strebel et al. (1989) suggest that in some cases, as for the chalk aquifer of eastern central England, a major increase in nitrate contamination occurred in the early 1970's but is now stabilized. This also seems to be the case in the eastern part of the United States (Smith et al. 1993). The data of Table 8 show a maximum value of nitrate accumulation rate in groundwater of 36 μM yr^{-1}.

The extent of groundwater affected by this increase is difficult to evaluate. A crude estimate of groundwater reservoirs by continents and hydrogeological zones was presented by UNESCO (1978). Taking into account the two uppermost zones, located above sea level and characterized by the most active water exchange, the average groundwater stock is estimated as 47 $\times$ 10^3 m^3 km^{-2} for Europe and 78 $\times$ 10^3 m^3 km^{-2} for North America. These values may seem low when compared with local estimates for important aquifers: in Europe, the 3 most important aquifers are those associated respectively with Permo-triassic sandstone, Cretassic chalk, and Tertiary sand

and gravels. These have stocks of water which range between 100 and 650 × 10^3 m^3 km^{-2} (Comm. European Communities 1982). An even higher stock (25 × 10^6 m^3 km^{-2}) is estimated for the upper Rhine alluvial aquifer. However, these highly productive aquifers cover less than 20% of the area of the continent.

Combining the estimates for the maximum nitrate concentration increase and the groundwater stock yields a maximum estimate of the rate of nitrogen storage in groundwater of intensively cultivated areas of the order of 25 kg N km^{-2} yr^{-1} for Europe and 40 kg N km^{-2} yr^{-1} for North America. This is only 1% of the net anthropogenic inputs of nitrogen to Europe and 3% for North America (Table 4). Groundwater storage may, however, be significant in some localized areas where high rates of increase in nitrate concentrations are observed in some large aquifers, as in the region of intensive agriculture in northwestern Europe. The sand aquifer of central Belgium is an example, with an estimated storage of nitrate of approximately 200 kg N km^{-2} yr^{-1}. The upper Rhine aquifer is another example.

Denitrification and storage of nitrogen in wetlands

Wetlands are important sites of denitrification and in some conditions can greatly reduce nitrogen movements through the landscape. For instance, in a study of an agricultural system in Maryland (USA), Peterjohn and Correll (1984) observed that nitrate discharged from a corn field in surface runoff and groundwater flow into a riparian forest was reduced by 90% before emerging into a stream. Unlike lakes where both accumulation of nitrogen in sediments and denitrification contribute significantly to nitrogen losses from the water (see next section on "Denitrification and Retention of N in Streams, Rivers and Lakes"), loss of nitrogen in wetlands is usually thought to be due largely to denitrification (Jansson et al. 1994; but see Brunet et al. 1994). Unfortunately, it is extremely difficult to evaluate overall nitrogen removal in wetlands at the scale of large regions, since the percentage nitrogen loss in wetlands varies with local physiographical, and hydrological conditions (Arheimer & Wittgren 1994). Also, with increasing area of wetlands in a watershed, nitrogen removal efficiency per area tends to decrease since the wetland area which first "sees" high nitrogen waters has the most impact (Arheimer & Wittgren 1994). Further, at least some wetlands seem to retain little if any nitrogen; in a study of 5 wetlands in Ontario, Devito et al. (1989) reported retentions ranging from −12% to +4% of inputs and not significantly different from zero. These wetlands transformed inorganic nitrogen into organic nitrogen, indicating the importance of measuring all nitrogen species to adequately determine nitrogen retention in wetlands. The complexity of nitrogen transformations within wetlands is further emphasized in the work of McClain et al. (1994) and Triska et al. (1994).

Strips of riparian wetlands between agricultural fields and streams can be particularly effective in removing nitrogen (Peterjohn & Correll 1984; Lowrance et al. 1984; Vought et al. 1994). However, often agricultural runoff reaches streams without passing through such buffers; for instance, even 25 years ago when fertilizer applications were relatively modest, more than half of the nitrate in surface waters in Illinois is estimated to have entered streams directly from agricultural drainage through drainage tiles (Kohl et al. 1971). Much agriculture in the North Atlantic basin occurs on former wetlands, and drainage is necessary to maintain agricultural productivity. This drainage not only facilitates nitrogen movement through drain tiles but has often removed the wetland characteristic of buffer strips between streams and agricultural fields (Vought et al. 1994). Krug (1993) notes that in southern Sweden, the conversion of the last 10–15% of land into agricultural use often has disproportionate effects on nutrient fluxes to aquatic systems since this involves removal of riparian fringes. He attributes up to half of nitrogen fluxes in streams to removal of these fringing wetlands.

Restoration and creation of wetlands offer mechanisms for potentially lowering nitrogen fluxes. Rosenberg et al. (1991) argue that wetland restoration, where feasible, is by far the cheapest alternative to lowering nitrogen fluxes in rivers. Haycock et al. (1993) urge the use of riparian wetland buffers along streams, particularly first and second order streams, as the most effective means of lowering nitrogen fluxes through the landscape.

Denitrification and retention of N in streams, rivers and lakes

Once in aquatic systems, nitrogen can be further lost through denitrification or storage in the sediments of lakes and reservoirs rather than delivered to the ocean. A number of budget studies which provide an estimate of nitrogen retention within the aquatic ecosystem portion of drainage basins are summarized in Table 9. These budgets consider edge-of-field exports of nutrient and point discharges as inputs to the river system. They therefore do not take into account nutrient retention by riparian buffer zones, discussed in the previous sections. The budgets do, however, include retention within lakes, ponds, and impoundments which are part of the aquatic continuum of the river systems considered.

In moderately loaded river systems, in-stream and in-river processes account for losses of approximately 10 to 20% of total nitrogen inputs. Nitrogen retention seems lower in rural or forested streams with total nitrogen inputs less than 1,000 kg N km^{-2} yr^{-1} per area of watershed, while it might amount to nearly 50% in heavily polluted rivers like the Scheldt (Table 9). Processes responsible for the observed nitrogen retention may include permanent deposition of particulate nitrogen in flood plains and impoundments, and denitrification either in the water column or in the sediments. Deposition of particulate nitrogen in the alluvial plain of large rivers during flood events or in reservoirs may be considerable: values as high as 10 to 100 g N m^{-2} yr^{-1} have been reported (Johnston et al. 1984; Billen et al. 1989). The significance of these processes for total nitrogen removal from river systems depends upon the existence and location of permanent deposition sites.

Denitrification in the water column occurs in case of severe oxygen depletion. This was and still is the case in some large, heavily polluted rivers of Western Europe, such as the Thames, the Scheldt, and the Seine Rivers, below major discharges of untreated sewage. Denitrification in such cases can sometimes eliminate all nitrate originating form upstream sources, such as leaching from agricultural soils. Paradoxically, improvement of sewage treatment can lead to increased nitrogen delivery to the ocean by rivers due to improved oxygen status and lowered rates of denitrification (Billen 1990; National Research Council 1993).

In well oxygenated water systems, which comprise the majority of rivers draining into the North Atlantic Ocean, benthic denitrification is the major process removing nitrogen. Some representative values of the observed rates of nitrogen removal by denitrification in lakes and river-bed sediments are shown in Table 10. Areal rates measured in stream beds are often higher than in lake sediments, probably because of a higher supply of organic matter in the streams. The net impact of benthic denitrification in streams versus lakes on the overall nitrogen budget of a river system depends upon a number of factors. Kelly et al. (1987) proposed a model of nitrogen removal through benthic denitrification in lakes with oxic hypolimnia based on simple mass balance calculations. Their model relates nitrogen loss through denitrification to water residence time (t), mean depth (z), and an average mass transfer coefficient (S_n, in m yr^{-1}) representing the height of the water column from which nitrate can be removed per unit time. S_n can be approximated by the ratio between mean areal denitrification rate and mean nitrate concentration in the water column, if first order kinetics are assumed for the rate of denitrification. For lakes, the data in Table 10 suggest S_n is typically in the range of 5 to 20 m yr^{-1}, which allows a prediction of nitrogen retention as shown in Figure 7. This is in very good agreement with the retention data deduced from budget studies in a number of lakes and impoundments, also shown in Figure 7, which range from 20 to 80% of inputs. To apply the model of Kelly et al. (1987) to river and streams, the data in Table 10 suggest S_n values in the range of 20 to 50 m yr^{-1}. The ratio between depth and residence time can be

Table 9. Nitrogen budgets for selected river basins. Rivers are listed in order of nitrogen inputs to the watersheds. Losses with rivers are calculated by the difference between measured inputs and exports; this includes losses within lakes, ponds, and impoundments in the watershed.

	Area (km^2)	Population density (km^{-2})	Inputs (kg N km^{-2} yr^{-1})	In-River Losses (kg N km^{-2} yr^{-1})	% N Loss in river	Reference
Viroin	554	50	650	0	0	De Becker 1986
Aare	8,096	119	735	145	19	Santschi et al. 1977
Seine	64,565	195	1,440	140	10	Billen et al. (unpubl)
Marne	12,730	91	1,710	145	8.5	Billen et al. 1994
Meuse	2,080	159	2,308	770	33	De Becker 1986
Dyle	275	279	2,690	190	7	De Becker 1986
Scheldt	16,200	426	4,490	2,045	45	Billen et al. 1985

Table 10. Rates of denitrification in the sediments of selected lakes and rivers.

	Denitrification (mmol m^{-2} hr^{-1})	Method*	Nitrate conc. (μM)	Sn (M yr^{-1})	Reference
Lakes					
Ontario lakes	0.01 to 0.025	c	7 to 20	5 to 13	Kelly et al. 1987
Lac du Der	0.3 to 0.7	a	100	10 to 25	Garnier et al. 1994
NL Reservoir	0.12 to 0.4	b	285	3.6 to 12	Admiraal et al. 1988
Bryup Langso	0.17 to 0.33	a,b	114	13 to 25	Anderssen 1977
Kvind so	0.10 to 0.25	a,b	71	12 to 30	Anderssen 1977
Rivers					
Dyle	2 to 4	a	500 to 600	30 to 70	De Becker 1986
Orneau	1	a	300	30	De Becker 1986
Meuse	0.9	a	150	52	De Becker 1986
Seine	0.5 to 3	a	150 to 300	30 to 87	Chesterikoff et al. 1992
Oise	1.6	a	210 to 285	50 to 67	Billen et al. 1989
Nottawasaga	0.2 to 0.6	a	150 to 350	5 to 35	Hill 1983
Carnagigue	0.15 to 0.27	d	200 to 650	2 to 12	Chatarpaul et al. 1980
Grachten	0.3 to 4.3	b	350	7.5 to 107	Van Kessel 1977
Swift's Brook	1.3	b			Kaushik and Robinson 1976
Gr.Ouse Trent	0.1 to 3.8	b			Owens et al. 1972
Ivel+Gade	0.3 to 4.3	b			Edwards & Rolley 1965
Potomac	0.21 to 0.24	e	100	18 to 21	Seitzinger 1987
Ontario streams	0.58	b	350	15	Wyer and Hill 1984

* Methods:

a. Net nitrate consumption measured with belljar or incubated cores;

b. Net nitrate consumption estimated from input-output balance;

c. Gross nitrate consumption measured by acetyelene blockage technique;

d. Gross nitrate consumption measured by ^{15}N technique;

e. Gross nitrate consumption measured by N_2 flux.

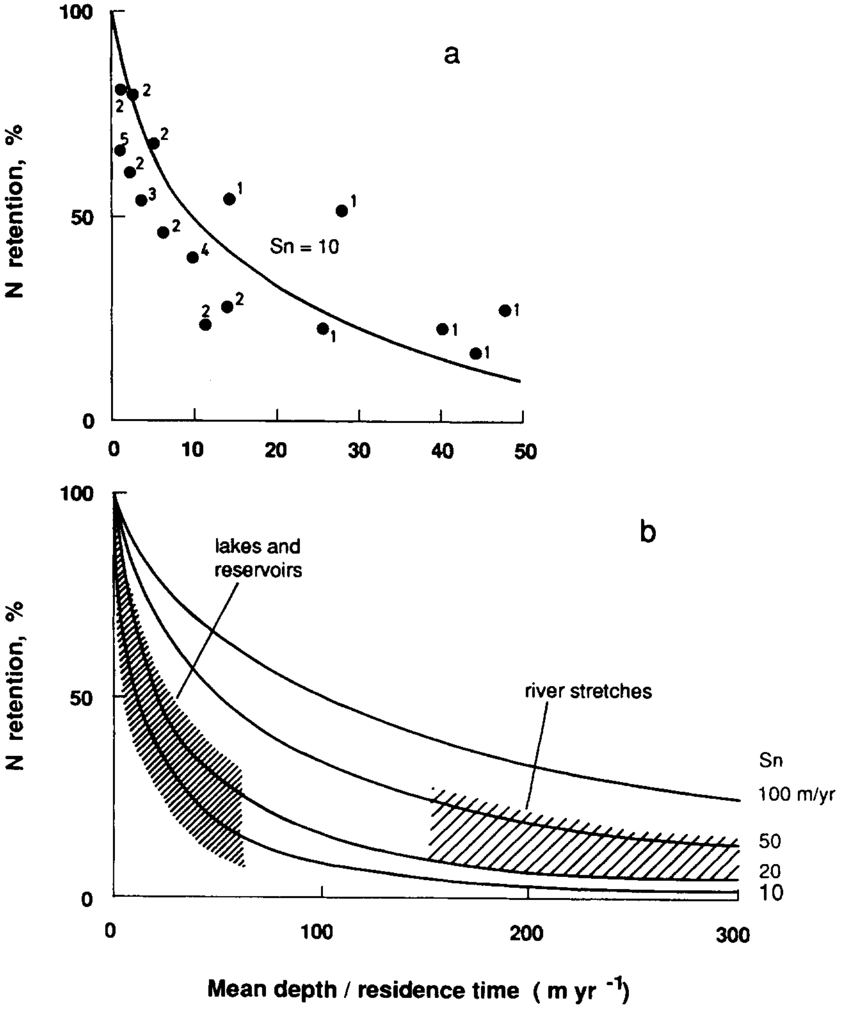

Figure 7. Kelly et al.'s (1987) model of nitrogen retention in unstratified water bodies; S_n is a mass transfer coefficient depending on benthic denitrifying activity and water column nitrate concentration; see text for model explanation.

a. Nitrogen retention expected for $S_n = 10$ m yr^{-1}, compared with observed data for a number of lakes; 1. Anderssen (1977); 2. Dillon & Molot (1990); 3. Garnier et al. (1994); 4. Calderoni et al. (1978); 5. Schelske (1985); 6. Galicka & Penczak (1989).

b. Nitrogen retention expected for a range of S_n values for the conditions representative of lakes and reservoirs and for streams and rivers in a typical river system of the North Atlantic basin.

calculated by stream order for a typical Atlantic river system. As depth and residence time both increase with increasing stream order, this ratio varies little and is typically in the range of 150 to 300 m yr^{-1}. We therefore predict that nitrogen retention in river channels is only 5 to 20% of inputs (Figure 7), in spite of higher denitrification rates, because of the much lower residence time characterizing river systems in comparison to lakes.

Reservoirs and lakes slow the movement of water and may, therefore, be sites of substantial nitrogen removal. For instance, reservoirs on the Nile and Colorado Rivers have average water residence times of 3.5 and 1.8 years respectively (van Der Leeden et al. 1990). However, most of the river basins feeding into the North Atlantic Ocean have few major reservoirs, and average water residence times are seldom increased by more than 0.33 years (van Der Leeden et al. 1990). Assuming that large reservoirs have average depths of at least 10–20 meters, it is highly unlikely that more than 20% of nitrogen inputs are removed in reservoirs in the regions draining into the North Atlantic (Figure 7). This is probably an overestimate, since most reservoirs in the regions draining to the North Atlantic Ocean are located on smaller order rivers relatively upstream in the watersheds. Areas very rich in lakes, such as the formerly glaciated Baltic and Northern Canada regions (Meybeck 1994, 1995), may show higher retention. Much of the nitrogen loading to rivers (particularly from agricultural and domestic sources) probably occurs downstream of most lakes and reservoirs.

Riverine fluxes in the temperate zone: Comparison to pristine conditions

What were nitrogen fluxes to the North Atlantic like prior to the widespread practice of agriculture and the industrial revolution? How can such fluxes be estimated? One approach is to assume that pristine watersheds, or the most pristine that can be found, represent the pre-agricultural and pre-industrial condition. Meybeck (1982) used this approach to estimate total riverine nitrogen flux to the world's oceans in the absence of human influence; he determined an average nitrogen export from relatively pristine watersheds of 355 kg km^{-2} yr^{-1}, corresponding to an average concentration (dissolved and particulate organic and inorganic nitrogen) of 67 μM (Table 11). Lewis (1986) compiled another set of data on "minimally disturbed" forested watersheds; he found N exports ranging from 84 to 998 kg km^{-2} yr^{-1}, corresponding to average total N concentrations of 5.9 to 82 μM (Table 11). Meybeck's (1982) reference level for pristine conditions, and at least two of the "minimally disturbed" sites compiled by Lewis (1986) – one temperate and one tropical – exhibit nitrogen exports per watershed area that are comparable to or greater than several of the regions in the North Atlantic basin (Figure 2a and Table 11).

Table 11. Concentration of total nitrogen (dissolved and particulate) and nitrogen export for "minimally disturbed" forested watersheds (after Lewis 1986, Lewis 1981, and Hedin et al., 1995).

	Total N (μM)	N export (kg km^{-2} yr^{-1})	N deposition (kg km^{-2} yr^{-1})	Old growth forest?
Venezuela	29.	998	745	Yes
West Africa	82.	126	1,910	??
Northwest Ontario (Canada)	24.	94	635	No
Oregon, USA	5.9	118	150	Yes
New Hampshire, USA	34.	401	880	No
Colorado, USA	14.	84	480	??
Chile, high mountains	11.*		<100	Yes
Chile, coastal	17.*		<100	Yes
Global average (Meybeck 1982)	67.**	355**		No

* dissolved forms only for Chilean study.

** calculated from Tables 2 & 6 of Meybeck (1982).

Several factors affect nitrogen export from forested watersheds, including the maturity of the forest (Vitousek & Reiners 1975; Emmett et al. 1993; Reynolds et al. 1994), the amount of nitrogen fixation (Binkley et al. 1992), and the amount of nitrogen deposition (Johnson 1992). Many forests which are typically considered pristine in fact have relatively high rates of nitrogen deposition from human activity, and therefore might be expected to export more nitrogen than would have been the case prior to the industrial revolution. Hedin et al. (1995) have demonstrated remarkable differences in nitrate concentrations in streams draining old-growth forests, and these differences seem related to differences in atmospheric deposition (but see Johnson 1992). Where deposition is low, as in some forests in Chile, nitrate concentrations are 3 or more orders of magnitude less than in forests in the northeastern U.S. receiving high levels of atmospheric deposition. We suggest that the best reference sites for export of nitrogen from pristine conditions would be old growth forests receiving very low rates of atmospheric deposition of nitrogen (Hedin et al. 1995). This approach is still problematic, as natural variations in the amount of nitrogen fixation and denitrification could cause nitrogen export from pristine old-growth forests to vary. Also, aggrading forests (which are more likely to retain nitrogen) occur naturally due to periodic forest fires. Further, in some parts of the North Atlantic basin, grasslands are a dominant feature of the undisturbed landscape, and may have high rates of both nitrogen fixation and denitrification.

We have several potential ways to estimate pristine conditions. The Andrews Experimental Forest in Oregon, USA, has a relatively low atmospheric deposition of nitrogen (150 kg km^{-2} yr^{-1}) and an export in stream waters estimated as 118 kg km^{-2} yr^{-1} (Table 11; Lewis 1986; Sollins et al. 1980). The northern Canadian region of our study is another relatively undisturbed region, and reflects a compilation of data that is consistent in scale and method with our other analyses. The value here is 76 kg N km^{-2} yr^{-1} (Figure 2a), and an independent study of several Canadian watersheds by Clair et al. (1994) arrived at an average export value of 79 kg N km^{-2} yr^{-1}. Estimates for fluxes in the Mackenzie and Lena rivers, two large relatively undisturbed rivers, are somewhat higher, averaging 230 kg N km^{-2} yr^{-1} (Meybeck, pers. comm.). Historical data on nitrogen export in the Connecticut River suggest a late 1800's value of 110 kg N km^{-2} yr^{-1} (Jaworski, unpublished). Finally, though not statistically significant, the y-intercept of our regression of nitrogen export in rivers against total net anthropogenic nitrogen inputs (Figure 5a) is consistent with the rest of these values at 103 kg N km^{-2} yr^{-1}.

This 3-fold variation in estimates of pristine inputs is probably very real, and in addition to spatial variation, pristine fluxes may have fluctuated both higher and lower in time. The detailed nitrogen budget for the Andrews Experimental Forest shows some of the pitfalls of assuming a uniformity among pristine sites in nitrogen inputs or outputs. For example, nitrogen fixation by epiphytic lichens at Andrews was estimated as 280 kg N km^{-2} yr^{-1} (Sollins et al. 1980), perhaps a low estimate of total nitrogen fixation as asymbiotic soil fixation on fallen logs was not measured. Thus, total inputs are similar to moderately impacted sites in low-elevation forests of the southeastern U.S. and upper New England, particularly if we add a small unmeasured dry deposition input. However, outputs of nitrogen from Andrews were still estimated to be significantly lower than total inputs (Sollins et al. 1980). Even in this old-growth site where carbon and nitrogen storage in vegetation is thought to be decreasingly slightly over time (- 280 kg N km^{-2} yr^{-1}), accumulation of nitrogen in fallen logs and in soil organic matter is probably substantial (780 kg N km^{-2} yr^{-1}), and the ecosystem is thought to be still accumulating nitrogen (Sollins et al. 1980).

Rates of nitrogen loss from eastern Canadian stands may be lower than rates from the Andrews Forest for several reasons: rates of nitrogen fixation may be lower, the stands may be aggrading more rapidly from more recent fire activity, and denitrification in lakes and rivers may be removing more nitrogen than in the Andrews system. Also, precipitation patterns certainly differ. Andrews has a very high annual precipitation and receives most of its rain during the cooler winter months when plant uptake may be low; retention of atmospherically-deposited nitrogen has been shown to be lower during the dormant season (Lajtha et al. 1995). It is reasonable to assume that at least some patches of forest in large old-growth stands have a positive net ecosystem production, either in recovering vegetation or in soil or woody detritus, but to quite varying degrees; successional history alone could cause several-fold differences in storage, and thus output rates.

Although the north Canadian region value is at the low end of the range for nitrogen export, it is interesting to note that this region has some fertilizer inputs and atmospheric deposition values that are well above pristine. On the other hand, the northern Canadian rivers region contains many lakes and has a relatively long surface water residence time, which would tend to lead to nitrogen losses through denitrification and sediment accretion in lakes; the surface water residence time on the Canadian shield is an order of magnitude larger than in most of the other regions of the North Atlantic basin (Meybeck 1994, 1995).

We therefore chose to use 76 to 230 kg N km^{-2} yr^{-1} as the range for our estimates of pristine river nitrogen flux from the temperate regions of the North Atlantic basin. This range suggests that human activity has increased nitrogen fluxes from temperate regions to the North Atlantic Ocean several fold (Table 12). In some regions, such as the northeast coast of the US and the North Sea, we estimate fluxes have increased 5 to 20 fold, while most of the other regions show 2 to 9-fold increases. In comparison, Larsson et al. (1985) using an independent approach of comparing northern Swedish river fluxes (assumed to be pristine) with other riverine inputs to the Baltic have estimated a 4-fold increase due to human influence, which is quite consistent with our analysis. We do not believe that the above range for pristine export should be used to estimate change in the tropical watersheds, for reasons discussed below in the section on "Nitrogen Balance in The Humid Tropics."

If instead of choosing this range of values as our pristine reference, we had instead used the compilation of Meybeck (1982), the natural pristine flux would be 355 kg N km^{-2} yr^{-1}, or 1.5 to 4 times larger than our pristine assumption. This would imply that many of the regions surrounding the North Atlantic have experienced relatively little change, and that in the case of Northern Canada, the rivers exported much more nitrogen to the ocean in their pristine state than they currently do. This is possible, if nitrogen sinks have increased in the watersheds or rivers, but we find it unlikely. We believe our analysis is the preferred one. We note that among rivers assumed to be relatively undisturbed by human activity, Meybeck (1982) included many which probably receive high atmospheric deposition of nitrogen. In addition, the Amazon is one of the rivers considered by Meybeck (1982) to be pristine, and as discussed below, pristine nitrogen fluxes from a tropical systems are likely to be larger, perhaps much larger, than those typical of the temperate zone.

Table 12. Estimated changes in river N export from temperate regions expressed as a ratio of current fluxes to a range of estimates of pristine fluxes. Our estimates of pristine fluxes (all in values of kg N km^{-2} yr^{-1}), ranged from 76 (the current flux from the Northern Canadian watersheds) to 230 (average value for Mackenzie and Lena Rivers). The values in the table reflect those two extremes. Other pristine estimates included an independent study of several Canadian watersheds (=79), the measured value at the Andrews Experimental Forest (=118), historical data from the Connecticut River (=110), and the y-intercept of the relationship between river N export and total anthropogenic N inputs (=103; Figure 4A). See text for full references and details.

Region	*Current river N export* "Pristine" river N export
North Canadian rivers	0.33 to 1
St. Lawrence basin	1.8 to 5.4
NE coast of US	4.7 to 14.1
SE coast of US	2.9 to 8.9
Eastern Gulf of Mexico	2.9 to 8.9
Mississippi basin	2.5 to 7.4
Western Gulf of Mexico	2.6 to 7.9
Baltic Sea	2.2 to 6.5
North Sea	6.3 to 19.1
NW European coast	5.6 to 17.1
SW European coast	1.6 to 4.8
North America	1.7 to 5.3
Europe	3.5 to 10.6

Nitrogen balance in the humid tropics

In comparison to many of the temperate North Atlantic watersheds, the Amazon and Orinoco basins appear minimally disturbed. Nitrogen deposition and fertilizer application are both relatively low in these regions (Tables 4 and 5; Figure 4), as is human population density (Figure 1a). As well, in sharp contrast to the temperate regions, these tropical regions show river nitrogen fluxes being roughly equal to the sum of anthropogenic NO_y deposition and fertilizer input (compare Figure 2a with Table 4). This might suggest a relatively pristine, steady-state nitrogen cycle, and in fact, Meybeck (1982) used the Amazon River as a baseline for estimating anthropogenic perturbations to riverine fluxes of carbon, nitrogen, and phosphorus in other watersheds. However, several lines of evidence indicate that the nitrogen cycle of the neotropics may be changing rapidly, and even the undisturbed nitrogen dynamics of many tropical systems may make them a misleading standard for measuring change in the rest of the world.

First, consider the unique aspects of the nitrogen cycle in many tropical forests. While nitrogen limitation of plant growth is widespread in temperate and boreal ecosystems (Vitousek & Howarth 1991), nitrogen is in relative abundance throughout much of the humid tropics. For example, N:P ratios in litterfall are around 11:1 in temperate and boreal forests; in the tropics they average approximately 23:1 (Vitousek 1984). This excess of available nitrogen leads to lower retention in plants and microbes, and therefore to a "leakier" terrestrial nitrogen cycle, where losses to both the atmosphere and aquatic systems may be high (Matson & Vitousek 1990). Tropical forests produce the highest reported fluxes of nitrous oxide from undisturbed systems (Keller et al. 1993). Total nitrogen fluxes within the forests of the Amazon are some 5 to 6-fold greater than those at the Andrews Forest (Bentley et al. 1982; Sollins et al. 1980; Jordan et al. 1982), an old-growth forest in the temperate zone with relatively low nitrogen deposition. Thus, even in a completely pristine world, nitrogen inputs from terrestrial ecosystems to rivers would probably be much higher in the humid tropics than in any other region, and therefore the use of the Amazon as an index of anthropogenic perturbations to temperate rivers may seriously underestimate the amount of change in the temperate region and globally.

In addition, the clearing and burning of forests in tropical regions may have a disproportionately large effect on regional nitrogen cycling. Combustion losses of nitrogen during conversion are typically around 50,000 kg km^{-2} (Ewel et al. 1981; Keller & Reiners 1994), and another 50,000 to 250,000 kg km^{-2} is lost over the following year or so (Berish 1983). Some of this "lost" nitrogen may be retained via anion exchange in deep soils (Matson et al. 1987), and some will be deposited onto nearby systems (Andreae et al. 1990), but land conversion clearly mobilizes large amounts of nitrogen. The 1980's rate of deforestation in the Amazon has been estimated at 1.5×10^4 km^2 yr^{-1} (Skole & Tucker 1993); if nitrogen losses from land conversion average 200,000 kg km^{-2} yr^{-1}, then deforestation is mobilizing approximately 3 Tg of nitrogen per year. This value is roughly equal to the annual river flux (Table 2), even though it occurs over only 0.25% of basin area. Thus, even though only about 6% of the basin has been cleared, the entire region may experience the effects.

On the other hand, the fraction of nitrogen mobilized by land conversion which eventually reaches the rivers is not clear, as both total nitrogen loss and the partitioning of those losses among various pathways can vary with climate, soil type and the methods of clearing. In one Costa Rican site, the dominant pathways for nitrogen loss appeared to be gaseous (Keller & Reiners

1994). In a neighboring region, Matson et al. (1987) showed that leaching dominated losses in the months following burning, but also showed that much of the nitrogen leached from surface soils was retained via anion exchange in deeper horizons. Such nitrate adsorption is not an important process in most temperate soils, but it can be substantial in the variable charge clays that are common to the humid tropics (Uehara & Gilman 1981), and therefore nitrogen which might appear to be lost from vegetation and surface soils might not actually enter hydrologic systems (Matson et al. 1987).

Land conversion alone is simply a mechanism for redistributing nitrogen within a region. Though it can substantially increase turnover times for otherwise recalcitrant pools of organic nitrogen, it does not provide new inputs, and therefore is unlikely to produce changes in aquatic nitrogen loading that are comparable to those brought on by the high nitrogen inputs to temperate systems. However, the subsequent urbanization or agricultural use of the converted land may lead to much higher nitrogen inputs to the region. Nitrogen deposition and fertilizer use are both increasing rapidly in equatorial latitudes (Galloway et al. 1995). Thus, the coming decades may see changes in tropical river and coastal systems similar to those already observed throughout North America and Europe.

Summary and conclusions

With currently available data, we are unable to fully account for the fate of anthropogenic nitrogen added to the temperate regions of the North Atlantic Basin. Nonetheless, some general conclusions are possible. Fertilizer and atmospheric deposition dominate total anthropogenic nitrogen inputs to the temperate regions as a whole, with fertilizer accounting for roughly two-thirds of the total. Rates of nitrogen export in rivers vary dramatically among the major regions, and are highly correlated with the amount of human-derived nitrogen applied to the landscape. Although export of N in rivers has clearly increased in many areas, all of the regions show that only a relatively small fraction of total human-derived nitrogen inputs can be accounted for in the rivers (roughly 25%). Most of the nitrogen added to regions through human activity is stored within the region or denitrified. Thus, it is critical to understand the other major controls over loss and/or storage of this anthropogenic nitrogen.

The overall nitrogen fluxes within and between the terrestrial, groundwater and river systems of the North Atlantic watershed are summarized graphically in Figure 8. Assuming that terrestrial primary producers take up some 5,000 to 15,000 kg N km^{-2} yr^{-1} in the North Atlantic basin as a whole (Duvigneaud & Denaeyer-DeSmet 1971), net external anthropogenic inputs of nitrogen represent as much as 7% to 22% of internal cycling. Fertilizer application dominates these inputs, but anthropogenic atmospheric deposition and nitrogen fixation associated with crop vegetation also contribute significantly. Nitrogen is transferred from terrestrial systems to the hydrosystem through soil leaching in both agricultural systems and forests and through direct wastewater discharge. Overall for the North Atlantic watershed, sewage and wastewater discharge only represents about 10% of total riverine delivery. Leaching from animal feedlots, which we have not quantified, may also be important. Storage of nitrogen in groundwater, while difficult to quantify, is probably at most a few percent of the rate of input of anthropogenically derived nitrogen. On the other hand, storage of nitrogen in forests may be significant, perhaps accounting for up to 26% of net anthropogenic nitrogen inputs to the temperate portions of the North Atlantic basin; the redistribution of ammonia from agricultural systems to forests through atmospheric transport may be important in the ability of forests to store this amount of nitrogen. Nonetheless, much of the net anthropogenic nitrogen input to the North Atlantic basin is not stored in forests or groundwater and does not flow to the oceans in rivers; by difference with values for the temperate areas, we calculate that on average at least 340 kg N km^{-2} yr^{-1} is probably denitrified or stored in wetlands and aquatic systems, or one third of net anthropogenic inputs to the temperate region. To the extent we have overestimated storage in forests, denitrification will be even more important. Denitrification in both wetlands and aquatic ecosystems is probably of importance.

This overall view of nitrogen cycling at the scale of the whole North Atlantic watershed masks differences among the watershed regions considered in this paper. Three extreme situations have been represented in Figures 9a, 9b, and 9c. The Amazon & Tocantins region (Figure 9a) is characterized by the lowest anthropogenic inputs of nitrogen. The relatively high nitrogen riverine delivery, with organic nitrogen being the dominant form, probably corresponds to the normal functioning of the tropical rain forest, characterized by high rates of natural nitrogen fixation, and phosphorus limitation of primary production; in a degree difficult to assess, deforestation may be impacting the Amazon riverine transport of nitrogen. Nitrogen fluxes from this region to the ocean actually exceed anthropogenic inputs of nitrogen.

The North Canadian region (Figure 9b) probably provides a better idea of what could have been the pristine nitrogen cycle in temperate regions. External anthropogenic nitrogen inputs and riverine delivery of nitrogen to the ocean both represent less than one fifth of the mean value for the overall North Atlantic when expressed on a per area basis. Export of nitrogen in rivers to the sea from this region represents only one third of the net anthropogenic nitrogen inputs, a value typical for many temperate regions in the North

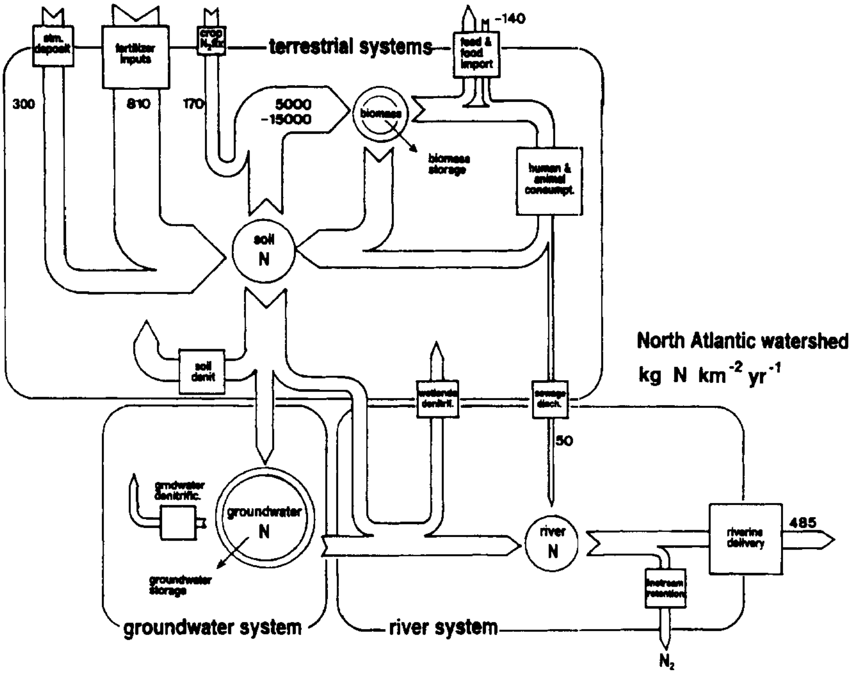

Figure 8. Schematic representation of nitrogen circulation within and between the terrestrial system, the groundwater system, and the river system of the North Atlantic basin as a whole. Values in kg N km^{-2} yr^{-1}. Width of arrows, although not strictly proportional, suggests the relative magnitude of the corresponding fluxes.

Atlantic basin. However, retention of nitrogen in aquatic ecosystems may be higher in this region than in many others because of the high residence time of surface waters (Meybeck 1994, 1995).

The North Sea watershed region (Figure 9c) shows the most perturbed situation, with a largely open nitrogen cycle. Fertilizer inputs (6,000 kg N km^{-2} yr^{-1}) completely dominate external inputs and represent more than one third of nitrogen uptake by the vegetation, resulting in an intense nitrogen soil leaching. The high population density causes an important discharge of sewage and wastewaters into surface water, roughly one third of the total riverine nitrogen flux. Although the rate of nitrogen export in rivers per area of watershed is about 20 times that of the North Canada region, it represents only 20% of total anthropogenic nitrogen inputs. The majority of nitrogen added through human activity is probably eliminated through denitrification, with a small amount stored in forests (Table 7).

The Mississippi basin and the northeastern U.S. coast also deliver respectively 7 and 14 times as much nitrogen per unit area as the North Canadian

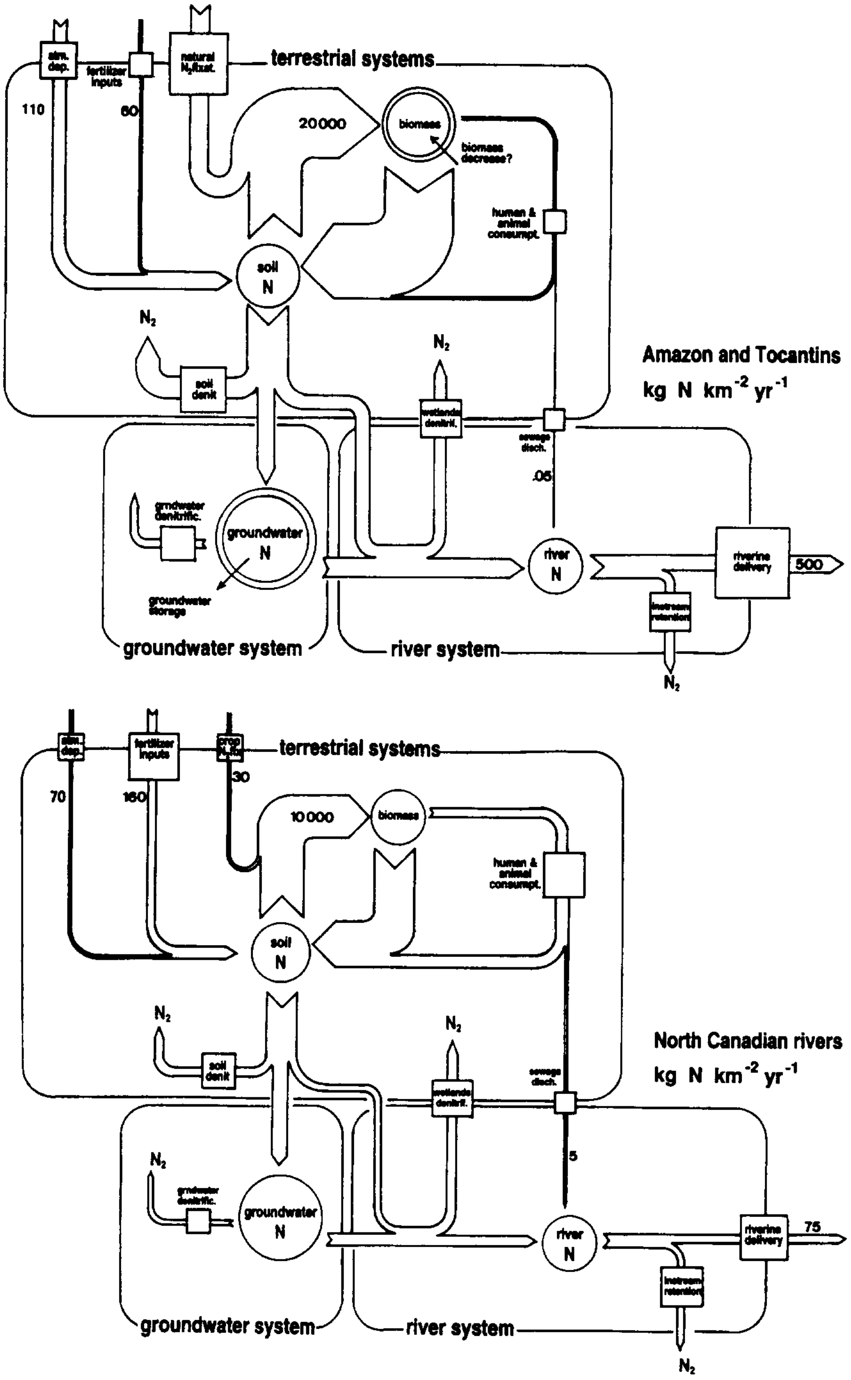

Figure 9. Schematic representation of nitrogen circulation in some watershed regions of the North Atlantic basin: a.) Amazon basin; b.) Northern Canadian rivers; c.) North Sea watershed.

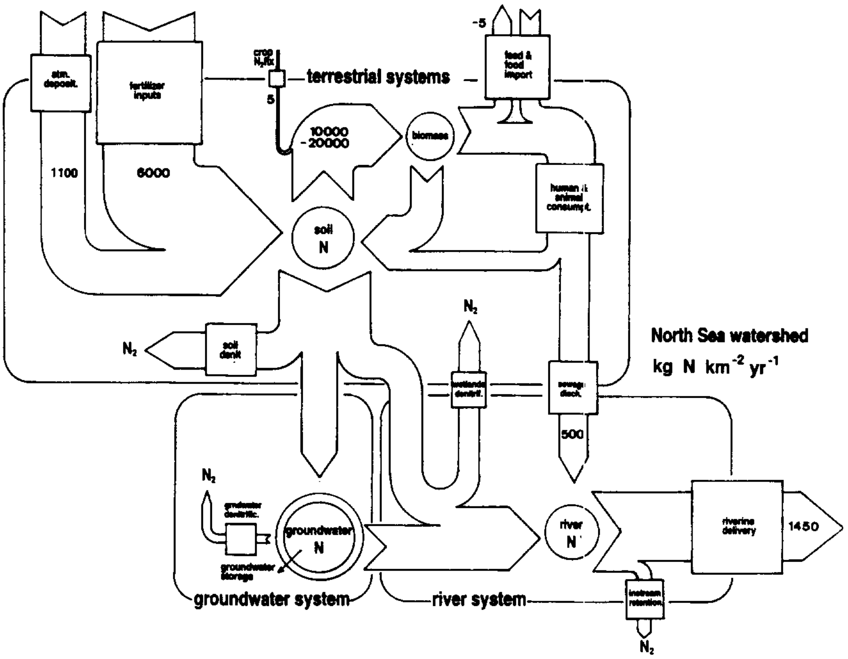

Figure 9. Continued.

region. They differ however from the North Sea basin region in the nature of the external anthropogenic inputs of nitrogen. The Mississippi basin is an intensive agricultural region, with moderate population density. Fertilizer inputs (1,840 kg N km^{-2} yr^{-1}) and nitrogen fixation by crop vegetation (1,055 kg N km^{-2} yr^{-1}) dominate the inputs, but as much as one third of these (1,300 kg N km^{-2} yr^{-1}) is exported as food and feed to other regions. The riverine nitrogen delivery to the coastal zone (565 kg N km^{-2} yr^{-1}) represents 25% of net anthropogenic inputs to the system. By contrast, the northeastern US coast region is characterized by high population density and limited agriculture, and imports of feed and food (1,000 kg N km^{-2} yr^{-1}) and anthropogenic nitrogen atmospheric deposition (1,200 kg N km^{-2} yr^{-1}) represent the two major external nitrogen sources. Inputs of nitrogen as fertilizer and crop N_2 fixation are also important, and the nitrogen delivery to the coastal sea (1,070 kg N km^{-2} yr^{-1}) is nearly twice that of the Mississippi basin.

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