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 atmo-

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

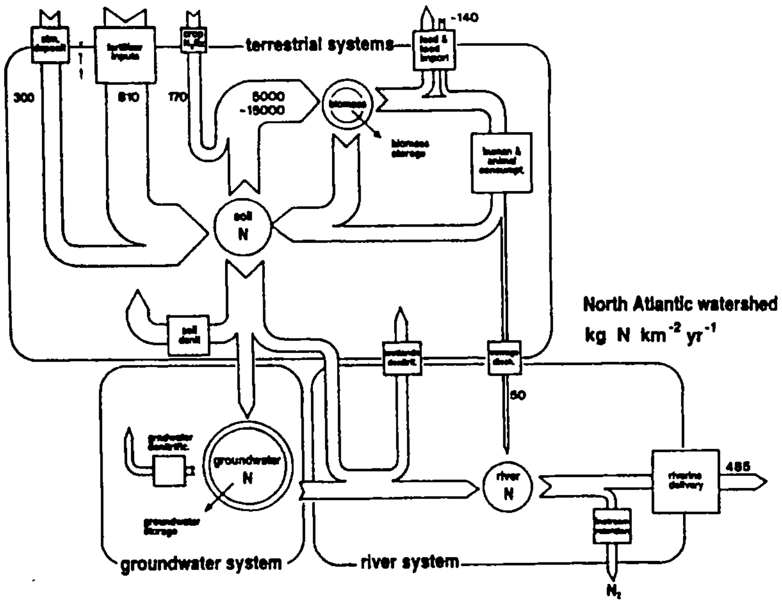

Figure 8. Schematic representation of nitrogen circulation within and between the terrestrial system, the groundwater system, and the river system of the North Atlantic basin as a whole. Values in kg N km^{-2} yr^{-1}. Width of arrows, although not strictly proportional, suggests the relative magnitude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

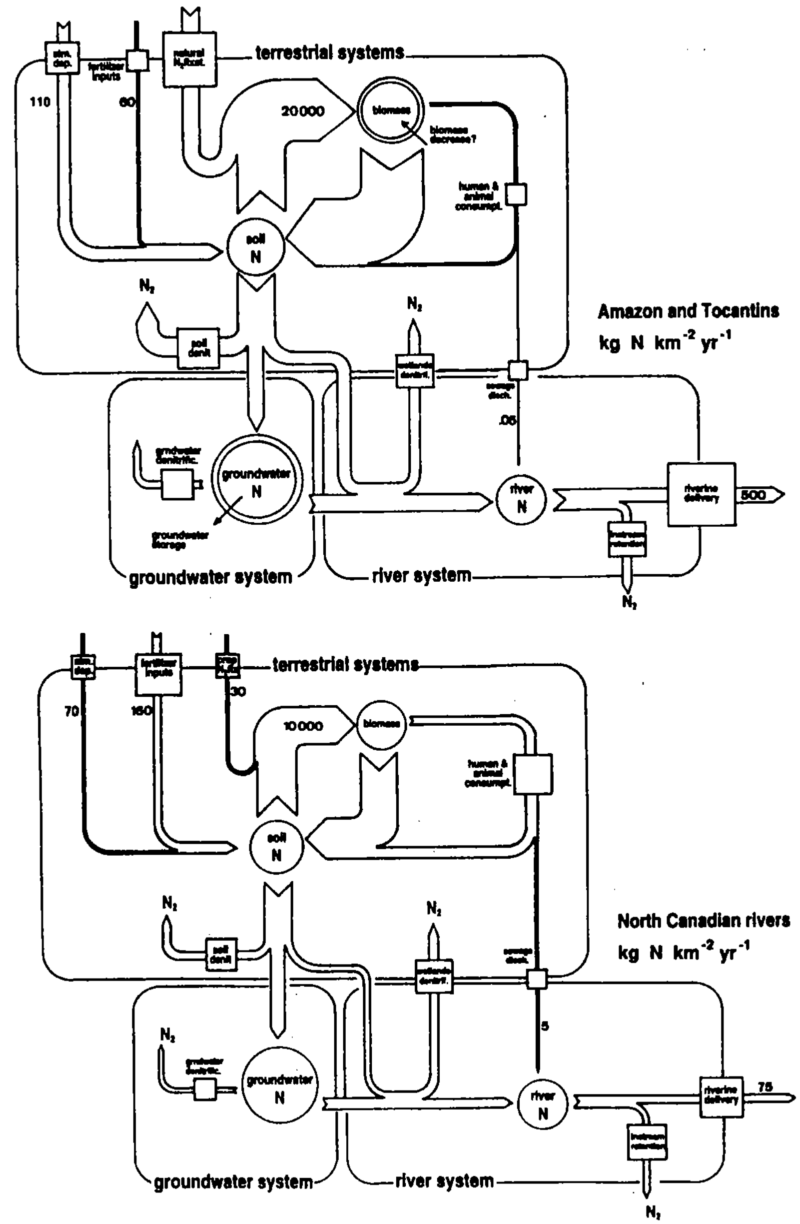

Figure 9. Schematic representation of nitrogen circulation in some watershed regions of the North Atlantic basin: a.) Amazon basin; b.) Northern Canadian rivers; c.) North Sea watershed.

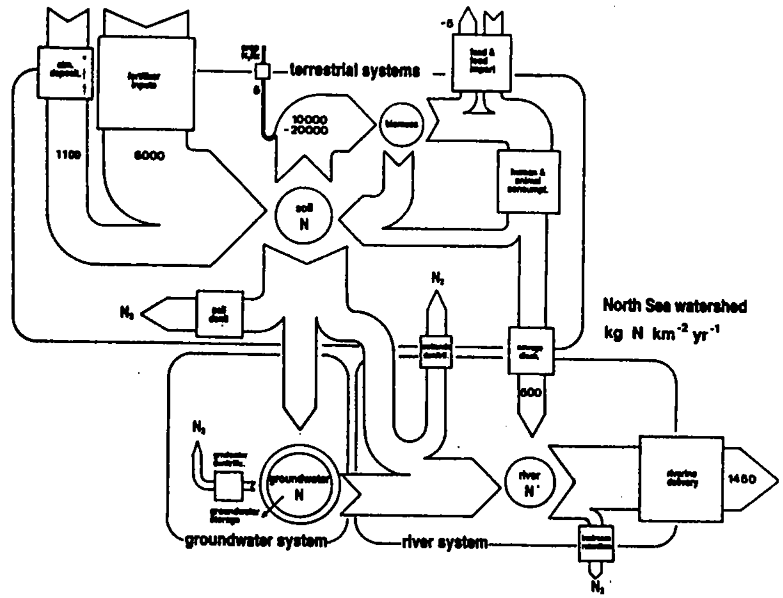

Figure 9. Continued.

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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Atmospheric Deposition of Nitrogen Oxides onto the Landscape Contributes to Coastal Eutrophication in the Northeast United States

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Recently compiled data document a 3–8-fold increase in nitrate fluxes from 10 watersheds in the Northeast United States since the early 1900s. During this period, nitrogen oxide emissions from combustion sources have increased about 5-fold. For 17 large watersheds with relatively minor agricultural or urban influences, riverine nitrogen fluxes from 1990 to 1993 were highly correlated with atmospheric deposition onto their landscapes and also with nitrogen oxide emissions into their airsheds. These relationships provided two methods of estimating riverine nitrogen export directly from either deposition or emission fluxes. For 10 benchmark watersheds with good historical data, about 36–80% of the riverine total nitrogen export, with an average of 64%, was derived directly or indirectly from nitrogen oxide emissions. Atmospheric deposition of nitrogen represented only about 25% of the airshed emissions with the remaining 75% transported out of the airshed. Nitrogen is the element most responsible for eutrophication in coastal waters of this region. Our analysis suggests a strong linkage between the increase in cultural eutrophication of the coastal waters of the Northeast United States and the increase in nitrogen oxide emissions from fossil fuel combustion.

Introduction

Eutrophication, caused by excessive inputs of nitrogen to rivers in the coastal zone, is one of the greatest factors altering water quality in estuarine ecosystems in the United States (*1*). Globally, human activity has accelerated nitrogen cycling and at least doubled the rate of nitrogen fixation over natural levels (*2, 3*). Several studies have documented increased nitrogen fluxes to estuaries, such as the Mississippi River mouth (*4*), Narragansett Bay (*5*), and the Baltic Sea (*6*). Recent comparisons of nitrogen fluxes from large regions around the North Atlantic Ocean suggested that human activity may have increased nitrogen fluxes to the coastal rivers of the Northeast United States by 5–14 times over natural rates (*7*). A recent study (*8*) showed that an increase in nitrogen loading not only caused a loss of terrestrial diversity but also increased the nitrogen export. At regional scales, a recent study (*9*) tested the hypotheses that riverine export of nitrogen increases as anthropogenic input of nitrogen increases and that net anthropogenic input onto the landscape is a good predictor of river export. Dramatic regional and global shifts in the spatial movement of nitrogen in agriculture has been examined as a result of the rise in cash crop production and intensive animal production (*10*).

In general, while we are reducing much of the uncertainty in estimates of the anthropogenic sources of nitrogen to the ~~landscapes of~~ watersheds, the quantitative linkages of the sources with the increased riverine fluxes remain poorly characterized. Human sewage, animal wastes, and runoff of fertilizer are usually assumed to dominate (*4, 11, 12*), although a significant contribution from atmospheric deposition has been suggested (*13–15*). Atmospheric releases and deposition of nitrogen have increased (*3*). This nitrogen deposition can contribute to downstream nitrogen fluxes in streams and rivers if the input exceeds the uptake needs of vegetation (*16*) or if the soils are particularly permeable and unable to hold on to the nitrogen (*17*). For the northeast U.S. region as a whole, atmospheric deposition of nitrogen onto the landscape, which originates mainly from fossil fuel combustion, exceeds all other individual nitrogen inputs, including fertilizer, leguminous crop fixation, and imported food and feeds (*7*). The regional scale analysis (*9*) reported that food and feed import was the largest source of nitrogen to the Northeast United States.

The major objectives of this paper are to (i) compile and document historical riverine nitrate fluxes for 10 benchmark watersheds that represent the landscape of the northeastern United States; (ii) explore spatial response relationships of riverine export fluxes of nitrogen with atmospheric deposition of nitrogen and with airshed nitrogen emissions for watersheds with minor agricultural and urban influences; (iii) estimate current and historical riverine nitrogen export as a result of nitrogen oxide emissions for the 10 benchmark watersheds directly from deposition and emission fluxes; (iv) compare, on a total nitrogen bases, the landscape anthropogenic inputs, riverine exports and landscape consumption for the 10 benchmark watersheds; and (v) reconstruct the 1900–1993 total nitrogen loading fluxes from atmospheric deposition, wastewater discharges, and agricultural runoff to the coastal waters of the northeastern United States.

Study Area

The Northeast United States study area encompasses the watersheds (north to south) of the Gulf of Maine including Massachusetts Bay, Buzzards Bay, Narragansett Bay, Long Island Sound, Hudson-Raritan Estuary, Delaware Bay, and Chesapeake Bay. The watersheds have a combined area of about 475 400 km^2 with an estimated 1990 population of 55 million people. About 90% of the 22 billion L/day of municipal and industrial wastewater is discharged directly into tidal coastal waters.

Ten large coastal watersheds, with sufficient historical water quality monitoring data, were selected as benchmarks (Table 1) to document the historical trends and to examine current nitrogen landscape loadings and riverine export fluxes. The ten watersheds have a drainage area of about 218,000 km^2 and a wastewater discharge flow of about 3.5 billion L/day. These 10 watersheds, which include the Penobscot River flowing into the Gulf of Maine; the Merrimack into Massachusetts Bay; the Connecticut into Long Island Sound; the Delaware and Schuylkill into Delaware Bay; and the Susquehanna, Potomac, Rappahhanock, and James Rivers flowing into Chesapeake Bay, uniformly span the northeastern United States study area.

The landscape of the 10 watersheds is about 70% forested, 25% agricultural, and 5% urban. Portions of the Potomac, Susquehanna, and Schuylkill Watersheds support intense crop and animal production resulting in large nutrient runoff fluxes.

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TABLE 1. Basic Data for 10 Benchmark Watersheds, North to South Array

watershed	USGS station no.	drainage area (km²)	airshed area (km²)	av river discharge ($m^3\ s^{-2}\ km^{-2}$)	wastewater flows (L/day) (10^6)	land use: forest (%)	land use: agriculture (%)	land use: urban (%)
Penobscot	1036390	20 109	155 500	0.02053	68	62	21	2
Merrimack	1100000	12 005	70 300	0.01987	492	91	5	4
Connecticut	1184000	25 019	86 400	0.02122	568	78	12	10
Hudson	1358000	20 953	270 680	0.01723	68	72	23	5
Delaware	1463500	17 560	265 400	0.01781	454	85	12	3
Schuylkill	1474500	4 903	170 400	0.01503	379	48	42	10
Susquehanna	1578310	67 314	251 500	0.01638	984	64	32	4
Potomac	1646580	29 940	313 300	0.01076	322	59	37	4
Rappahannock	1668000	4 134	41 600	0.01068	15	54	44	2
James	2035000	16 206	518 600	0.01415	129	75	23	2
average		21 814	214 368	0.01637		69	25	5

The Connecticut, Delaware, Merrimack, and Schuylkill Watersheds have the highest wastewater flows per unit area of watershed.

Data Sources and Methods

As part of a regional analysis of nitrogen inputs to the North Atlantic Ocean, data previously were compiled to estimate riverine nutrient and mineral fluxes for 33 major watershed monitoring sites between Maine and Virginia for the period 1990−1993 (*7*, *18*; Jaworski et al., in preparation). Riverine nutrient and mineral export fluxes were estimated by multiplying the average annual concentration of a specific nutrient or mineral by the mean annual river discharge. Except for nitrates, these 4-year average annual estimates were usually within ±5% of the estimates in the Chesapeake Bay watersheds that were conducted by the U.S. Geological Survey using a load estimation model (*19*). While the 4-year average annual estimates for nitrates were usually 5−15% lower than the flux predicted by the USGS model, the estimates were well within annual variations, which were often a factor of 2−3.

Nitrate and stream flow data used to calculate the nitrate fluxes for 10 benchmark watersheds (Figures 1a and 2b) come from drinking water monitoring programs (unpublished reports from municipal drinking water authorities), from the Massachusetts State Health Agency monitoring program from 1890 to 1915, and from the published U.S. Geological Survey Water Resource Data annual reports. Both river and drinking water sampling data are included and cover most of this century. The annual summary data reflect mainly monthly sampling; however, some of the more recent sampling has been reduced to bimonthly.

To assess the quality of the early data, comparisons were made with aperiodic water quality data obtained by the U.S. Geological Survey (*20*) between 1905 and 1920 and by the Massachusetts State Health Agency from 1890 to 1915. The recent drinking water data were also compared with recent U.S. Geological Survey data. In general, there was very good agreement where data sets overlapped. For continuous data sets, such as for the Potomac Watershed, the nitrate, chloride, and sulfate concentrations increased steadily over time, further confirming that the increases in chemical concentration observed were not due to major changes in analytical methodology. Large changes in concentration occurred as river discharge increased or, for example, during the 1960−1970 drought. Chlorides and sulfates, being conservative, went up in concentration while nitrates, being non-conservative, went down in concentration during the drought period.

For the ten benchmark watersheds, historical nitrate fluxes (Figures 1a and 2b) were calculated from the nitrate concentration and the historical river discharge data. The current inorganic, organic, and total nitrogen (TN), historical nitrate export fluxes, which were extrapolated to a common historical base year (1900), and total organic carbon (TOC) fluxes are presented in Table 2.

In our review of the water quality data from a large number of monitoring sites in the study area, some of these riverine systems had significantly elevated nutrient levels and thus were clearly impacted by large wastewater discharges and/or by extensive agricultural fertilizer and animal waste runoff. To determine whether riverine nitrogen exports were related to atmospheric deposition of nitrogen, watersheds were screened to eliminate those that were heavily affected by wastewater discharges, and agricultural runoff were identified. A total of 55 monitoring sites, for which there were nutrient flux estimates for the 1990−1993 period, were screened.

Elevated phosphorus and potassium levels were used as indicators of significant increases in nutrients from wastewater discharges and/or agriculture runoff. High levels of ammonium, which is the unoxdized form of nitrogen, indicate the presence of significant wastewater and/or agriculture runoff that has not been nitrified. Sites that were excluded were those that had phosphorus concentrations greater than 0.075 P mg L^{-1}, potassium concentrations above 2.0 K mg L^{-1}, or ammonium nitrogen concentrations greater than 0.06 N mg L^{-1}. Watersheds that had large impoundments on their main stems were also excluded since such impoundments will lower riverine nitrogen export fluxes by encouraging denitrification (*7*).

This screening left 17 monitoring sites with low phosphorus and potassium concentrations and for which urban wastewater discharges and agricultural runoff should be relatively small. Low to moderate application rates of commercial fertilizer and animal waste in the 17 watersheds, as obtained from a national watershed-based analysis (*12*), further suggest that the agricultural practices would not be the major source of nitrogen in these watersheds and thus have only a minor affect on the export fluxes of nutrients. While it would be ideal to have all 17 watersheds with the same percent of the landscape in agricultural production so that land use would not be an additional unknown variable, the intensity of crop and animal production and not the percent of agricultural land appears to be the most important factor.

The final selected, minimally impacted by urban wastewater and agricultural runoff, 17 monitoring sites had rather large watersheds ranging in size from 120 to 20 109 km², with an average area of 6286 km² (Table 3). About 74% of the landscape of the 17 watersheds was forested, 22% was in agriculture, and 4% was urban. While the 10 benchmark watersheds had a slightly higher percent agricultural landscape at 25%, the average riverine TN export for the 10 was significantly higher at 766 kg of N $km^{-2}\ yr^{-1}$ (Table 2) as compared to 422 kg of N $km^{-2}\ yr^{-1}$ for the 17 watersheds (Table 4). Small sub-basins in the 10 benchmark watersheds with intense agricultural practices often had riverine TN export fluxes of 2000−3000 kg of N $km^{-2}\ yr^{-1}$.