

## Nitrogen and phosphorus budgets of the North Atlantic Ocean and its watershed

J. N. GALLOWAY<sup>1</sup>, R. W. HOWARTH<sup>2</sup>, A. F. MICHAELS<sup>3</sup>,  
S. W. NIXON<sup>4</sup>, J. M. PROSPERO<sup>5</sup> & F. J. DENTENER<sup>6</sup>

<sup>1</sup>*Environmental Sciences, University of Virginia, Charlottesville, VA 22903 USA;* <sup>2</sup>*Ecology and Systematics, Cornell University, Ithaca, NY 14853 USA;* <sup>3</sup>*Bermuda Biological Station for Research, St. Georges GE-01 Bermuda;* <sup>4</sup>*Graduate School of Oceanography, University of Rhode Island, Narragansett RI 02882 USA;* <sup>5</sup>*RSMAS, University of Miami, Miami, FL 33149 USA;* <sup>6</sup>*Department of Airquality, Wageningen, NL 6700 EV Wageningen, The Netherlands*

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**Abstract.** Anthropogenic food and energy production extensively mobilize reactive nitrogen (N) in the watershed of the North Atlantic Ocean (NAO). There is wide spread N distribution by both hydrologic and atmospheric processes within the watershed of the NAO, resulting in reactive N accumulation in terrestrial systems. Net denitrification in most estuaries and continental shelves exceeds the amount of N supplied to the shelves by rivers and requires a supply of nitrate from the open ocean. Thus riverine N is only transported to the open ocean in a few areas with the flow from a few major rivers (e.g., Amazon). Atmospheric N deposition to the open ocean has increased and may increase the productivity of the surface ocean. In addition, as a consequence of increased Fe deposition to the open ocean (due in part to anthropogenic processes), the rate of biological N-fixation may have increased resulting in N accumulation in the ocean. Phosphorus (P) is also mobilized by anthropogenic processes (primarily food production). Relative to N, more of the P is transported across the shelf to the open ocean from both estuaries and major rivers. There are several consequences of the increased availability of N and P that are unique to each element. However, the control on primary productivity in both coastal and open ocean ecosystems is dependent on a complex and poorly understood interaction between N and P mobilization and availability.

### Introduction

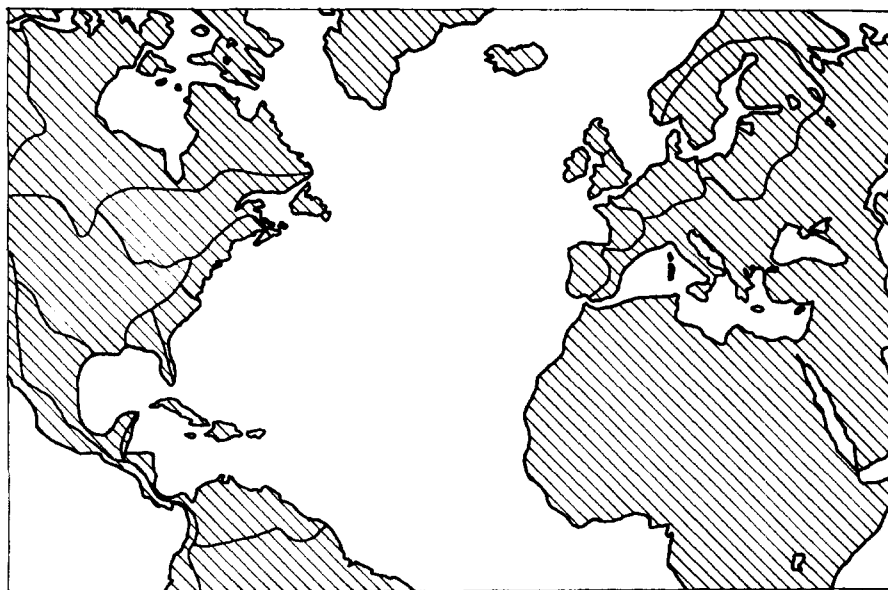
Nitrogen (N) is a key element of many biogeochemical processes and can be a limiting element of aquatic and terrestrial ecosystem processes (Schlesinger 1991; Vitousek & Howarth 1991). However, about 99% of global N exists as stable atmospheric N<sub>2</sub> (Mackenzie et al. 1993) and thus is unavailable to ecosystems unless it is converted into a reactive N species (reactive N = NH<sub>3</sub>, NH<sub>4</sub><sup>+</sup>, organic N, NO, NO<sub>2</sub>, HNO<sub>3</sub>, NO<sub>3</sub>, N<sub>2</sub>O<sub>5</sub>, HNO<sub>4</sub>, HNO<sub>2</sub>, NO<sub>2</sub><sup>-</sup>, and NO<sub>3</sub><sup>-</sup>). Once created, one species of reactive N can be converted into other species of reactive N by a variety of chemical and microbial processes. In addition, reactive N species are very mobile via atmospheric and hydrologic pathways. After formation, reactive N can only be converted back to unreactive N<sub>2</sub> by denitrification, an anaerobic process which only occurs

at significant rates in specific types of ecosystems. Reactive N accumulates in the environment if the denitrification rate is less than the rate of N-fixation.

In the absence of humans, natural processes create reactive N by biological N-fixation and lightning. Biological N-fixation occurs in specific microbes when atmospheric  $N_2$  is converted to  $NH_3$  by the enzyme nitrogenase. Lightning produces NO by the reaction of  $N_2$  and  $O_2$  at high temperatures. The former process is about two orders of magnitude greater than the latter on a global basis (Galloway et al. 1995). Human intervention in the N cycle has increased the formation rate of reactive N by fertilizer production, legume and rice cultivation, and combustion of fossil fuels. Results from several recent analyses of the global N cycle (Mackenzie et al. 1993; Ayres et al. 1994; Galloway et al. 1995) generally agree that anthropogenic activities mobilize about  $10 \text{ Tmol N yr}^{-1}$  ( $\text{Tmol} = 10^{12}$  moles) and that human activities mobilize N at rates equal to natural terrestrial processes.

There is significant distribution of anthropogenic N by hydrologic and atmospheric transport. Combustion of fossil fuels injects reactive N (NO) directly into the atmosphere. Fertilizer and cultivation N increase the productivity of agricultural landscapes. However, on average, no more than 50% of the applied N fertilizer is removed by crop harvest; the remainder is lost to the atmosphere or hydrosphere, or stored in the soil (Howarth et al. this volume). In addition, N fixed in crops has a short residence time; it is quickly transformed into human or animal waste, which also results in significant inputs to the atmosphere and hydrosphere (Howarth et al. this volume). If the N introduced into the environment by fossil fuel combustion, fertilizer production and cultivation is not denitrified, then reactive N accumulates in downwind or downstream ecosystems.

The mobilization, distribution and accumulation of anthropogenic N impacts a number of physical and ecosystem processes. As discussed in Howarth et al. (this volume), Nixon et al. (this volume), and Michaels et al. (this volume), increased availability of reactive N increases forest productivity, and as a consequence stores atmospheric  $CO_2$ , contributes to forest decline (if soil is N-saturated) and climate change, and results in shifts in community structure and ecosystem function. It also increases coastal eutrophication in estuaries (including their wetlands), and results in increased N supply to oligotrophic mid-ocean gyres with concomitant effects on the ecology of the upper ocean. In addition to being important to ecosystems, reactive N also affects atmospheric chemistry (Prospero et al. this volume). High levels of  $NO_x$  ( $NO + NO_2$ ) play an important role in the photochemical production of  $O_3$  (Moxim et al. 1994).  $NH_3$  is a major source of alkalinity in the atmosphere and a source of acidity in soils (Schlesinger 1991). Although  $N_2O$  is not viewed as a reactive form of N in the troposphere, it adsorbs IR



*Figure 1.* The North Atlantic Ocean and its watershed. The watershed is subdivided into its drainage basins. The boundary between the shelf and the open ocean is 200m depth.

radiation and acts as a greenhouse gas. In the stratosphere,  $N_2O$  impacts  $O_3$  concentrations (Warneck 1988). Thus, any change in the rate of formation of reactive N (or  $N_2O$ ), its global distribution, or its accumulation rate can have a fundamental impact on many environmental processes.

Our understanding of the global distribution, fate and impacts of anthropogenic N is insufficient because of a lack of data. To partially alleviate this problem, it is necessary to examine the N cycle on a smaller scale where data are more available, while still keeping the salient features of the N cycle – natural and anthropogenic sources, atmospheric and hydrologic transport processes, and agricultural, forested, freshwater, coastal and marine ecosystems. Hence we focus on a watershed analysis in general, and the North Atlantic Ocean and its watershed (NAO&W) specifically (Figure 1). We selected the watershed scale because its physiographic and hydrological features shape a biogeochemical system; these same features shape systems of human interaction. We selected the NAO&W because, compared to other ocean basins, it contains a rich data record of the atmosphere: watershed: ocean units on N cycling and because of the high degree of N disturbance by human activities.

Our analysis of the NAO&W N budget was centered about a workshop, held in May 1994 at Block Island, RI, USA. The central scientific question of the workshop was:

What are the current sources and sinks of nitrogen in the North Atlantic Ocean and its watershed? How might these have been changed over natural background levels as a result of human activity?

Since the impact of anthropogenic N on ecosystems is determined in part by the relative availability of N and P, our investigation of the N budget of the NAO&W includes an analysis of the magnitude and fate of P transferred from continents to the ocean.

To address the question of N cycling on the scale of an entire ocean basin and its associated watershed, a unique group of individuals assembled, each with expertise in physical, chemical and biological aspects of marine, coastal, freshwater and terrestrial ecosystems, the atmosphere, and the connections among the components. Working groups were formed on the basis of reservoirs: atmosphere, watershed, coastal and shelf region, and open ocean. Each working group developed an annual N cycle for their respective reservoir. The watershed of the NAO and associated coastal segments were further divided (Figure 1) to investigate spatial variability in N and P fluxes. The task of each group was to characterize the internal N fluxes and the hydrologic/atmospheric export losses for their reservoir; input fluxes to the reservoir were the domain of the upstream or upwind group. To enhance integration, each group was allowed 'to borrow' members from other groups to address specific issues of N exchange.

The primary products of the workshop are the working group papers on the N and P fluxes of each sub-reservoir: atmosphere – deposition to watershed and oceans (Prospero et al. this volume); watershed – N inputs, fates, and riverine losses to the coastal margins, and riverine P fluxes to the coastal ocean (Howarth et al. this volume); coastal and shelf region–fate of riverine and atmospheric inputs (Nixon et al. this volume); and the open ocean (Michaels et al. this volume). In addition, in the process of developing these papers, it became apparent that several topics warranted special attention. Thus accompanying these papers are four notes:  $\text{NH}_3$  exchange between the atmosphere and the NAO (Quinn et al. this volume), N-fixation in the NAO (Lipschultz and Owens, this volume); denitrification in the ocean boundaries (Seitzinger and Giblin, this volume); and shelf areas of the North Atlantic Ocean (Pilson and Seitzinger, this volume).

This paper is a synthesis of the workshop findings. It presents the N and P budgets for the entire NAO&W system by addressing the following specific topics:

- Mobilization of reactive N in the NAO&W
- Re-distribution of reactive N among the subsystems of the NAO&W
- Removal of reactive N from the NAO&W relative to sources
- P distribution patterns in the NAO&W

- Anthropogenic N and P impacts on the NAO&W

An advantage of examining the NAO&W as one system is that it provides an overview; a disadvantage is that it masks the extensive spatial variability in the N and P budgets of the portions of four continents, and associated coastal margins, that constitute the watershed of the NAO. This variability is extensive. Europe and North America have significantly greater increases in the mobilization of reactive N than do Africa and South America. We highlight some of the detailed regional findings from the 14 sub-watershed portions of the watershed of the NAO and adjacent coastal regions and refer the readers to the appropriate papers for the rest of the detail.

## The results

We illustrate the overall behavior of N and P in the NAO&W by dividing the NAO&W into five systems: atmosphere, watershed ( $26.9 \times 10^6 \text{ km}^2$ ), estuaries, shelf (to 200m depth,  $5.7 \times 10^6 \text{ km}^2$ ), and open ocean ( $100 \times 10^6 \text{ km}^2$ ) (e.g., Figure 2). Shaded arrows in Figure 2 represent reactive N creation; open arrows represent reactive N transfers between the NAO&W sub-systems. The arrow width is sized to the median flux magnitude. The ranges about the median fluxes are found in the text. Units are  $10^9 \text{ moles yr}^{-1}$  ( $\text{Gmol yr}^{-1}$ ). For consistency, the fluxes in this paper have been rounded to two significant figures from the values reported in the accompanying papers.

### *Mobilization of reactive N in the NAO&W*

There are two sources of reactive N – creation within, and transport to, the NAO&W. Reactive N is created and mobilized by biological N-fixation, lightning, commercial N fertilizer use, fossil fuel combustion and legume cultivation. The watershed and open ocean are the most important source regions for reactive N in the NAO&W; per area, the land masses of the North Atlantic are a far greater source of reactive N, reflecting the greater human disturbance on land (Figure 2).

*Watershed.* Fertilizer use introduces  $1,600 \text{ Gmol yr}^{-1}$  to the watershed of the NAO and is the most important anthropogenic N source, followed by the deposition of  $\text{NO}_y$  ( $\text{NO}_y = \text{NO}_x + \text{HNO}_3 + \text{NO}_3^- \text{ aerosol} + \text{other oxygen-containing N species except N}_2\text{O}$ ) from fossil fuel combustion ( $590 \text{ Gmol yr}^{-1}$ ), and legume cultivation ( $330 \text{ Gmol yr}^{-1}$ ). The total anthropogenic N addition to the NAO watershed is  $2,500 \text{ Gmol yr}^{-1}$  (Howarth et al. this volume). Figure 2 shows a watershed input of anthropogenic  $\text{NH}_x$  ( $\text{NH}_3 + \text{NH}_4^+$ ) from atmospheric deposition ( $290 \text{ Gmol yr}^{-1}$ ). We view this input as

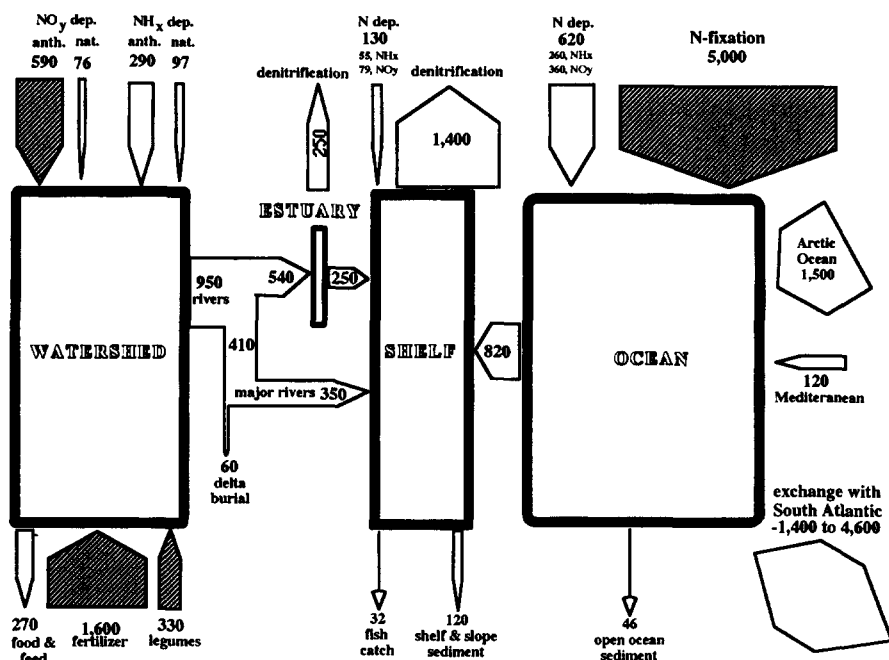


Figure 2. The N budget for the North Atlantic Ocean and its watershed ( $\text{Gmol yr}^{-1}$ ). The stippled arrows represent the introduction of new sources of reactive N into the NAO&W. The open arrows represent exchange of reactive N between the subsystems of the NAO&W.

an internal recycling of  $\text{NH}_x$  following its introduction by fertilizer use and subsequent  $\text{NH}_3$  volatilization from soils and animal waste (Howarth et al. this volume).

There are two natural sources of reactive N: lightning and terrestrial biological N-fixation. Lightning sources are small relative to fertilizer sources – the global total is only about  $210 \text{ Gmol yr}^{-1}$  (Levy et al. 1996). Natural terrestrial biological N-fixation for the watershed of the NAO may be important relative to anthropogenic N sources. Globally, the range of fifteen estimates is 5,700 to 13,600  $\text{Gmol yr}^{-1}$  (as reviewed in Jaffe 1992; Mackenzie et al. 1993). Thus, natural biological N-fixation in the watershed of the NAO, with 18% of the earth's land area, could be significant. However, given the probable strong latitudinal dependence on the N-fixation rate, the dearth of data for many tropical and temperate ecosystems, and our focus at examining the fate of anthropogenic N mobilized in the watershed of the NAO, we choose not to estimate natural terrestrial biological N-fixation rate. We do point out however, that studies more specifically focused on developing an integrated N cycle for watershed units need to include natural biological N-fixation.

The relative importance of anthropogenic N sources varies by sub-watershed unit (as defined in Figure 1). Agricultural N sources dominate inputs in many regions, particularly the Mississippi and the North Sea sub-watersheds. In the NE United States,  $\text{NO}_y$  deposition as a consequence of fossil fuel combustion is the major control over river N. In the temperate region, there is a strong correlation of total N flux with population density. This feature is manifested in a strong linear correlation between river fluxes of total N and the sum of anthropogenic N inputs (Howarth et al. this volume). Although on the average, N in food and feed is exported from the watershed of the NAO (Figure 2), for some sub-watersheds it can be a significant source. For example, in the NE coast region, about 1/3 of the N sources are from food and feed import (Howarth et al. this volume).

*Estuary and shelf.* Biological N-fixation certainly occurs in estuarine ecosystems but its magnitude is small relative to riverine input of reactive N previously created in the NAO watershed. In the shelf region biological N-fixation is estimated to also be small, on the order of  $20 \text{ Gmol yr}^{-1}$  (Nixon et al. this volume).

*Ocean.* Biological N-fixation in the open ocean is the most important N source for the NAO&W. Michaels et al. (this volume) identify a source of nitrate of 3,700 to 6,400  $\text{Gmol yr}^{-1}$  (median, 5000  $\text{Gmol yr}^{-1}$ , Figure 2) within the main thermocline of the Sargasso Sea that they infer is caused by N-fixation. This N-fixation may be regulated by the supply of Fe in dust and thus may have occurred at a lower rate before the expansion of the Sahara in the early 1970s. The most conspicuous N-fixing organisms in the North Atlantic Ocean are the filamentous cyanobacteria *Trichodesmium sp.* Lipschultz and Owens (this volume) conclude that the measurements of N-fixation by *Trichodesmium sp.* imply a rate of only 1,100  $\text{Gmol N yr}^{-1}$  for this taxa alone. The difference between Lipschultz and Owens and Michaels et al. may come from the scarcity of the data, questions about the accuracy of the methods and from the potential contributions of other organisms than *Trichodesmium sp.*

The second general source of N to the NAO is the transport into the region of reactive N created elsewhere. The large scale currents in the NAO result in significant horizontal advection of nutrients (Michaels et al. this volume). There is strong northerly surface transport in the Gulf Stream. There is a net southerly flow in the North Atlantic Deep Water, and net northerly transport in the Antarctic Bottom and Intermediate Waters. Michaels et al. summarized a number of techniques to estimate meridional transport in the NAO. From the north they estimate that 300 to 2,800  $\text{Gmol yr}^{-1}$  (median: 1,500  $\text{Gmol}$

$\text{yr}^{-1}$ , Figure 2) are advected into the NAO from the Arctic Ocean. In addition, Michaels et al. (this volume) estimate that 50 to 200  $\text{Gmol yr}^{-1}$  (median: 120  $\text{Gmol yr}^{-1}$ , Figure 2) are transported from the Mediterranean to the NAO. To the south, the uncertainty of the calculation is such that both the magnitude and direction of transport are unknown. The range is  $-1,400 \text{ Gmol yr}^{-1}$  out of the NAO to the South Atlantic Ocean (SAO), to 4,600  $\text{Gmol yr}^{-1}$  from the SAO to the NAO. The implications for the overall balance of N in the NAO are discussed in a following section.

In summary, the watershed of the NAO receives its anthropogenic reactive N primarily from fertilizer use, with smaller contributions from combustion of fossil fuels and legume cultivation. Some regions within the watershed of the NAO, especially within the temperate zone, are heavily impacted by not only fossil fuel combustion and fertilizer application, but also N importation in foods and feeds. The NAO receives its reactive N primarily from N-fixation, although transport from the SAO may also be important. As discussed in the next two sections, additional N sources are atmospheric and hydrologic transport of reactive N created in the watershed.

#### *Re-distribution of reactive N among the subsystems of the NAO&W*

*Via atmospheric transport.*  $\text{NO}_x$  and  $\text{NH}_x$  are emitted into the atmosphere by natural and anthropogenic processes. The most important anthropogenic processes are energy production ( $\text{NO}_x$  emissions from fossil fuel combustion) and food production ( $\text{NH}_3$  emissions from fertilizer use and animal waste) (Prospero et al. this volume). Once emitted,  $\text{NO}_x$  and  $\text{NH}_3$  are transformed, transported downwind and deposited as  $\text{NO}_y$  and  $\text{NH}_x$ , respectively. Unlike ecosystems, there are no important atmospheric reactions that convert  $\text{NH}_x$  to  $\text{NO}_y$  and visa-versa. Thus deposition of anthropogenic  $\text{NO}_y$  and  $\text{NH}_x$  can be generally linked to energy and food production, respectively.

Total (present-day natural and anthropogenic)  $\text{NO}_y$  atmospheric deposition to the watershed, shelf and open ocean regions of the NAO&W is 670  $\text{Gmol yr}^{-1}$ , 79  $\text{Gmol yr}^{-1}$  and 360  $\text{Gmol yr}^{-1}$ , respectively (Figure 2) (Prospero et al. this volume; Howarth et al. this volume; Nixon et al. this volume; Michaels et al. this volume). Anthropogenic  $\text{NO}_y$  deposition (590  $\text{Gmol yr}^{-1}$ ) to the entire watershed of the NAO is about seven-fold greater than deposition of naturally produced  $\text{NO}_y$  (76  $\text{Gmol yr}^{-1}$ ) (Figure 2). In regions heavily impacted by fossil fuel combustion (i.e., NE coast of US),  $\text{NO}_y$  deposition is estimated to have increased by a factor of 30 relative to natural rates. For more remote portions of the watershed of the NAO (i.e., Amazon and Tocantins basin)  $\text{NO}_y$  deposition has increased about 2–3 times over natural values (Howarth et al. this volume).



A similar situation exists for  $\text{NH}_x$  deposition. The watershed, shelf and open ocean regions receive total deposition of  $390 \text{ Gmol yr}^{-1}$ ,  $55 \text{ Gmol yr}^{-1}$  and  $260 \text{ Gmol yr}^{-1}$  respectively (Figure 2). For the watershed of the NAO, current  $\text{NH}_x$  deposition is about 3 times greater than natural rates for the entire watershed and up to 12 times greater for regions of intense agricultural activity (i.e., NW European coast) and about two times for more remote regions (i.e., Amazon and Tocantins basin) (Howarth et al. this volume). Relative to riverine sources, direct N atmospheric deposition to estuaries is small (Nixon et al. this volume).

While we have not estimated the natural and anthropogenic components of N deposition to the shelf and open ocean portions of the NAO, there is likely to be a strong human influence, especially for  $\text{NO}_y$ , given the disparity between natural  $\text{NO}$  atmospheric sources and anthropogenic. As discussed in Prospero et al. (this volume) and Galloway et al. (1995), the present-day global deposition rate of  $\text{NO}_y$  is about five times greater than pre-industrial times largely due to emissions from energy production and biomass burning. The relative increase will be even greater for the NAO because it is adjacent to region of intense fossil fuel combustion. While there is no doubt that  $\text{NH}_x$  deposition to the NAO has increased (Prospero et al. this volume), the magnitude of the increase is more difficult to estimate because of a potentially large natural emission of  $\text{NH}_3$  from the ocean (Quinn et al. 1988). At this stage, the data on  $\text{NH}_3$  emissions for the NAO are too sparse to make an estimate of an ocean wide flux (Quinn et al. this volume). It should be noted that our estimates of total N deposition to the NAO&W may be underestimates for a growing literature suggests that atmospheric deposition of organic N species may be equivalent to deposition of  $\text{NO}_y$  and  $\text{NH}_x$ , especially in marine regions (Prospero et al. this volume).

There is significant spatial variability in the  $\text{NO}_y$  and  $\text{NH}_x$  deposition rates. By way of illustration, we examine  $\text{NO}_y$  (Dentener & Crutzen 1993) and  $\text{NH}_x$  (Dentener & Crutzen 1994) deposition across transects of  $45^\circ \text{ N}$  and  $5^\circ \text{ N}$  at ten degree blocks of latitude and longitude (Figure 3). To show how N deposition to the NAO compares to the North Pacific Ocean, we begin the transects at  $175^\circ \text{ W}$ . To show how European and African emissions impact deposition to the east of the study area, we extend the transect to  $35^\circ \text{ E}$ .

At  $45^\circ \text{ N}$ , N deposition in North America and Europe is about 20 and 30 times, respectively, greater than the Pacific Ocean (Figure 3). Downwind of North America there is a rapid reduction in  $\text{NO}_y$  and  $\text{NH}_x$  deposition to the NAO. However, even in the middle of the NAO region, N deposition is still about double that of the North Pacific Ocean. These model results support previous statements that N deposition to the entire NAO has increased as a consequence of human activity (Whelpdale & Galloway 1994; Prospero et

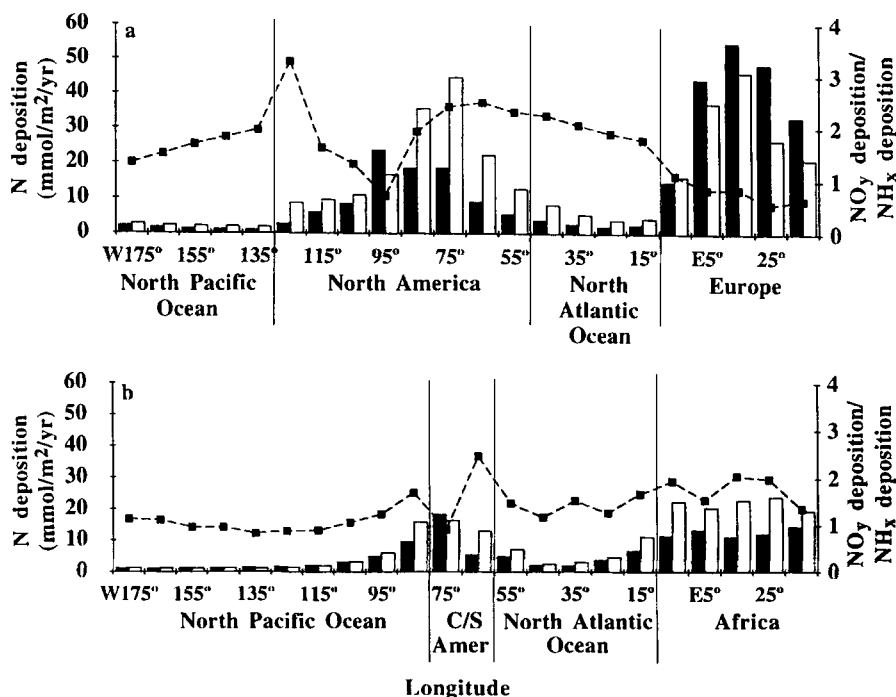


Figure 3.  $\text{NO}_y$  (open bars) and  $\text{NH}_x$  (solid bars) deposition ( $\text{mmol/m}^2/\text{yr}$ ) from  $175^\circ \text{W}$  eastward to  $35^\circ \text{E}$  at (a)  $45^\circ \text{N}$  latitude and (b)  $5^\circ \text{N}$  latitude. Ratio of  $\text{NO}_y/\text{NH}_x$  deposition shown by dotted line.

al. this volume). At the western boundary of Europe, N deposition increases significantly and remains high well outside of our study region due to atmospheric transport of European N emissions. Within the NAO&W study region, the relative importance of  $\text{NO}_y$  to  $\text{NH}_x$  deposition varies by about a factor of five. At the western boundary,  $\text{NO}_y/\text{NH}_x$  is about 0.7 reflecting the predominance of agricultural sources of  $\text{NH}_3$ . To the east, the ratio increases to about 2 due to the relative increase in energy over food production. The ratio gradually decreases over the NAO and is about 1 at the western boundary of Europe. It then decreases to about 0.5, again due to the strength of the agriculture sector relative to the energy sector.

For the  $5^\circ \text{N}$  transect, total deposition is significantly less than at  $45^\circ \text{N}$  (Figure 3) due to the reduced impact of  $\text{NO}_x$  and  $\text{NH}_3$  sources. Nonetheless, N deposition to the NAO at  $5^\circ \text{N}$  is still about 2–3 times greater than to the North Pacific Ocean. In addition, because of the predominant easterly winds, atmospheric transport of N is from east to west, accounting for the gradual reduction in deposition to the west of Africa and Central and South America. With the exception of the eastern portion of the Central and South America,

the  $\text{NO}_y/\text{NH}_x$  ratio varies between about 1 and 2, reflecting the importance of combustion over agriculture as N sources to the atmosphere.

In summary, there is a net transfer of  $\text{NO}_y$  and  $\text{NH}_x$  from the continental to the marine atmosphere. A substantial fraction of the  $\sim 2,500 \text{ Gmol yr}^{-1}$  of reactive N mobilized in the watershed of the NAO by human activities is deposited into the marine environment following atmospheric emission and transport.

*Via hydrologic transport.* N is hydrologically exchanged between the sub-systems of the NAO&W by riverine, estuarine and cross shelf processes.  $2,500 \text{ Gmol yr}^{-1}$  of anthropogenic N are introduced into the watershed of the NAO.  $950 \text{ Gmol yr}^{-1}$  of natural and anthropogenic N are lost via rivers to the marine environment;  $530 \text{ Gmol yr}^{-1}$  from the temperate portion of the watershed and  $420 \text{ Gmol yr}^{-1}$  from the tropical (Howarth et al. this volume). Since we do not have estimates of the natural N-fixation rate in the watershed, or precise estimates of the anthropogenic N in rivers, we can only say that the maximum loss of anthropogenic N via rivers in the temperate portion is 35%. Any amount of natural N-fixation within the watershed will reduce this percentage. The data are too sparse to make an equivalent estimate for the tropical portion.

Of the  $950 \text{ Gmol yr}^{-1}$  lost via rivers from the entire watershed of the NAO, about  $410 \text{ Gmol yr}^{-1}$  are from large rivers that debouch directly into the NAO shelf region; the remainder ( $540 \text{ Gmol yr}^{-1}$ ) flow into estuaries. Ultimately, large rivers and estuaries deliver about  $350 \text{ Gmol yr}^{-1}$  (290 from major rivers to shelf waters and  $\sim 61 \text{ Gmol yr}^{-1}$  to slope sediments) and  $170\text{--}340 \text{ Gmol yr}^{-1}$  to the NAO shelf region, respectively ( $250 \text{ Gmol yr}^{-1}$ , Figure 2) (Nixon et al. this volume). From the ocean side of the shelf region, about  $820 \text{ Gmol yr}^{-1}$  are transported to the shelf from the open ocean, primarily as nitrate (Nixon et al. this volume; Michaels et al. this volume). Thus the shelf region, via hydrologic transport, receives about  $1,400 \text{ Gmol N yr}^{-1}$  of N from major rivers, estuaries and the open ocean (Figure 2). It also receives  $\sim 20 \text{ Gmol yr}^{-1}$  by N-fixation (not shown on Figure 2). This amount, combined with the atmospheric flux of  $130 \text{ Gmol yr}^{-1}$ , results in an input of  $\sim 1,600 \text{ Gmol yr}^{-1}$  to the shelf. As discussed in the following section, most of the N delivered to the shelf region is denitrified.

There is significant spatial variability in the degree of hydrologic N transport. About two-thirds of the riverine N is injected from the western portion of the watershed of the NAO, about equally divided between North America and Central and South America. 25% of the N comes from one basin – the Amazon (Howarth et al. this volume). Similarly, all the major rivers that inject N directly to the shelf region are in the western portion of the watershed of the

NAO and again, the Amazon exerts a large influence. It accounts for about half of the  $600 \text{ Gmol yr}^{-1}$  of riverine N that are injected on to the NAO shelf (Nixon et al. this volume).

There is also spatial variability in the impact that human activities have had on hydrologic transfer of N between the NAO and its watershed. Using data from relatively pristine areas as an index of change, Howarth et al. (this volume) estimate that riverine N 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. However, we do not know the relative contribution of natural vs. anthropogenic activities. Deforestation may be contributing to the Amazon N flux, but tropical regions may also have naturally greater riverine fluxes due to higher amounts of N-fixation and phosphorus rather than N limitation of net primary productivity in forests (Howarth et al. this volume).

For the NAO shelf as a whole, N fluxes from rivers and estuaries ( $600 \text{ Gmol yr}^{-1}$ ) exceed atmospheric deposition ( $130 \text{ Gmol yr}^{-1}$ ) by about 4-fold (Figure 2), but this varies widely among regions of the shelf. For example, on the U. S. Atlantic shelf and on the NW European shelf, atmospheric N deposition may exceed estuarine exports.

### *Removal of reactive N from the NAO&W relative to sources*

This section discusses the removal of reactive N from the NAO&W by denitrification, storage in long term reservoirs (e.g., forests, sediments, groundwater), transport out of the NAO&W via the atmosphere, the hydrosphere and food and feed. It concludes with a summary of the N budget for the NAO&W (Table 1).

*Denitrification within the NAO&W.* The conversion of reactive N to  $\text{N}_2$  by biological denitrification decreases the reactive N stock in the NAO&W and can be viewed as a permanent sink (atmospheric  $\text{N}_2$  residence time is  $\sim 10^6$  yrs). Important locations of watershed denitrification are wetlands and the sediments of freshwater aquatic ecosystems. Howarth et al. (this volume) estimate that about  $>920 \text{ Gmol yr}^{-1}$  are denitrified or stored in wetlands, lakes, streams and rivers of the temperate portions. The equivalent value for tropical portions is unknown. As pointed out in Howarth et al. (this volume), there is significant spatial variability in the amount of denitrification and storage over the watershed of the NAO.

*Table 1. Summary of Nitrogen Fluxes for the Watershed, Estuary, Shelf and Open Ocean Regions of the North Atlantic Ocean and its Watershed, Gmol yr<sup>-1</sup>\**

INPUTS		OUTPUTS	
<b>• Temperate Watershed</b>		<b>• Temperate Watershed</b>	
Fertilizer (2)	1460	River discharge (2)	520
Combustion (2)	510	Food/feed (2)	230
N-fixation by legumes (2)	330	Groundwater storage (2)	<60
Natural biological N-fixation	??	Forest storage (2), [when nat. biolog N-fix is low]	<530
N-fixation by lightning	small	Denitrification & storage in wetlands, streams, rivers (2)	>920
Total input	2300	Total output	2300
<i>(w/o natural biological N-fixation)</i>		<i>(Plus whatever is fixed in natural biological N-fixation)</i>	
<b>• Tropical Watershed</b>		<b>• Tropical Watershed</b>	
Fertilizer, excl. Africa (2)	85	River discharge (2)	420
Combustion, excl. Africa (2)	74	Food/feed (2)	??
N-fixation by legumes (2)	??	Groundwater storage (2)	??
Food & feedstock imports	??	Forest storage (2)	??
Natural biological N-fixation	??	Denitrification & storage in wetlands, streams, rivers (2)	??
N-fixation by lightning	??	Total output	??
Total input	??		
<b>• Estuary</b>		<b>• Estuary</b>	
Rivers (2)	540	Transport to shelf (3)	250
Atmospheric deposition (1)	small	Storage in sediment(3)	small
Nat. biological N-fixation (3)	small	Denitrification (3, 5)	250
Total input	540	Total output	500
<b>• Shelf</b>		<b>• Shelf</b>	
From estuaries (3)	250	Denitrification (3, 5)	1400
Major rivers (2)	410	Delta, shelf, slope burial (3)	180
Atmospheric depositon (1)	130	Fish catch (3)	32
Nat. biological N-fixation (3)	20		
From ocean (3,4)	820		
Total input	1630	Total output	1612
<b>• Ocean</b>		<b>• Ocean</b>	
Nat. biological N-fixation (4,6)	5000	Denitrification (4)	small
Atmospheric depositon (1)	620	Storage in sediment (4)	46
From Arctic Ocean (4)	1500	To SAO (4)	??
From Mediterranean Sea (4)	120	Transport to shelf (3, 4)	820
From SAO (4)	??		
Total input	7240	Total output	966

\* The values in this table are averages and are presented to give an appreciation of the magnitude of the fluxes. For some parameters, the range around the average can be substantial. For all values, see paper for details. The numbers in parentheses refer to the original paper that discusses the flux: (1) Prospero; (2) Howarth; (3) Nixon; (4) Michaels; (5) Seitzinger and Giblin; (6) Lipschultz.

There are three other locations where denitrification may be important: estuaries, shelf and open ocean. Estuaries receive about  $540 \text{ Gmol N yr}^{-1}$  via rivers, of which about half is buried or denitrified, primarily the latter mechanism (Nixon et al. this volume). The shelf region receives about  $1,600 \text{ Gmol yr}^{-1}$  from major rivers, estuaries, atmospheric deposition, N-fixation, and the open ocean. It is estimated that  $\sim 90\%$  is denitrified in the shelf sediments and the water column; the remainder is buried in shelf and slope sediments or removed in fisheries harvest (Figure 2) (Nixon et al. this volume; Seitzinger & Giblin, this volume). Denitrification in the open ocean can occur in the sediments and the water column. Most of the sediment denitrification occurs in the shelf, which we have already accounted for. Open ocean denitrification is uncertain, but thought to be small because it requires areas with strong sub-surface oxygen minimum zones and anoxic basins. Anoxic conditions only occur in a few marginal seas like the Cariaco trench. Even if there is active denitrification in anoxic microenvironments within the water column (e.g. inside particles) then oceanic rates of denitrification must be small relative to the N-fixation rate. Denitrification removes nitrate and lowers the nitrate:phosphate ratios below the nominal Redfield stoichiometries of 16N:1P. In most of the NAO, nitrate:phosphate ratios are anomalously high, relative to the Redfield ratio (Fanning 1989; 1992; Michaels et al. 1994) indicating net N-fixation (Michaels et al. this volume).

*Storage within the NAO&W.* Storage in the watershed of the NAO occurs in biomass and groundwater. The temperate watershed of the NAO stores up to  $530 \text{ Gmol yr}^{-1}$  of the  $2,500 \text{ Gmol yr}^{-1}$  in forest biomass in an unknown mixture of anthropogenic and natural N; storage in groundwater of the temperate portion is less than a few percent ( $<60 \text{ Gmol yr}^{-1}$ ) (Howarth et al. this volume). Equivalent values for the tropical forest and groundwater are unknown (Howarth et al. this volume). Storage of reactive N in deltas and estuarine shelf and open ocean sediments is thought to be small; in all cases estimated N storage is  $<10\%$  of the N inputs (Figure 2) (Nixon et al. this volume; Michaels et al. this volume). The overall balance of fluxes in the open NAO indicates that it may be accumulating N at a rate of  $0.06\text{--}0.4\%$  per year, even given the considerable uncertainty in the transport estimates (Michaels et al. this volume). This apparent accumulation is largely a result of the large inputs from the estimated nitrogen-fixation and a modest atmospheric deposition, both of which may vary with time and are likely larger today because of human activity. There is a net flux of nitrate to the shelves to meet the shelf denitrification demand that exceeds the nitrogen supply from the rivers. In the pre-industrial world, these riverine N fluxes would also have been lower and the denitrification demands on oceanic nitrate larger. Unfortunately, the large

size of the total nitrate pool in the oceans and our short observational record preclude the direct observation of changes in the total stock of reactive N.

*Transport out of the NAO&W.* There are three transport mechanisms that distribute anthropogenic N beyond the borders of the NAO&W: atmospheric, hydrologic and food/feed exports. About 10% of the anthropogenic N formed in the watershed of the NAO is exported from the NAO&W as food and feed ( $270 \text{ Gmol yr}^{-1}$ , Figure 2) (Howarth et al. this volume). Hydrologic transport of N out of the region can occur via deep water flow to the South Atlantic Ocean. There is a potential for transport of up to 20 to 40% of the N fixed by biological activity in the NAO. The uncertainty in this transport term is such that there is also the potential for the SAO to be a source for N that is equivalent to N-fixation in the NAO (Figure 2) (Michaels et al. this volume). Atmospheric transport of N out of the region does occur; however, the magnitude is uncertain. Regions of significant transport are to the east of Europe and the west of Central and South America (Figure 3).

*N Budget of the NAO&W.* In summary, there are three important N sources to the NAO&W: anthropogenic N introduction to the watershed, N-fixation in the open ocean and transport of N into the NAO from adjacent water bodies. Natural biological N-fixation in the watershed of the NAO is probably an important, but unknown N source. Important N sinks are storage within the NAO&W subsystems, denitrification, and possibly transport to the SAO.

Significant uncertainties in some of the sinks and sources precludes an overall budget for the NAO&W. However, we can compare N sources and sinks for the NAO subsystems (Table 1). For the NAO temperate watershed,  $2,300 \text{ Gmol N yr}^{-1}$  are introduced by fertilizer, fossil fuel combustion and legume cultivation. Natural biological N-fixation is unknown but probably significant. Of this N,  $520 \text{ Gmol yr}^{-1}$  are lost via rivers; a small amount ( $<60 \text{ Gmol yr}^{-1}$ ) is stored in groundwater. Food and feed exports account for another  $230 \text{ Gmol yr}^{-1}$ . Storage in temperate forests is less than  $530 \text{ Gmol yr}^{-1}$ , storage and denitrification in temperate wetlands, streams and rivers is greater than  $920 \text{ Gmol yr}^{-1}$ . We are not able to differentiate between storage and denitrification in wetlands, streams and rivers. For tropical regions we have uncertain estimates of anthropogenic inputs and thus we are not able to estimate either forest storage or storage and denitrification in freshwater ecosystems (Table 1). It is clear, nonetheless, that the tropics export more N in rivers as a consequence of anthropogenic activities (Howarth et al. this volume).

The estuaries of the NAO&W receive most of their N from riverine discharge ( $540 \text{ Gmol yr}^{-1}$ ); atmospheric deposition and N-fixation are less

important. About half of the N delivered to estuaries is denitrified and half is transported to the shelf waters (Table 1).

The NAO shelf receives about  $1,600 \text{ Gmol N yr}^{-1}$  from: the ocean ( $820 \text{ Gmol yr}^{-1}$ ), major rivers ( $410 \text{ Gmol yr}^{-1}$ ), estuaries ( $250 \text{ Gmol yr}^{-1}$ ), atmospheric deposition ( $130 \text{ Gmol yr}^{-1}$ ), and N-fixation ( $20 \text{ Gmol yr}^{-1}$ ). Most of this N is denitrified, and only a small amount ( $<10\%$ ) is lost to delta, shelf and slope sediments ( $180 \text{ Gmol yr}^{-1}$ ) or as fish catch ( $32 \text{ Gmol yr}^{-1}$ ) (Table 1).

The major N source to the NAO is biological fixation ( $5,000 \text{ Gmol yr}^{-1}$ ) (Table 1). Hydrologic transport from the SAO could be very important, but both the sign and the magnitude of the transport is very uncertain. Estimates range from a loss of  $1,400 \text{ Gmol yr}^{-1}$  to the SAO to a gain of  $4,600 \text{ Gmol yr}^{-1}$  from the SAO. In spite of the large uncertainties in all the fluxes, the NAO appears to be accumulating N in today's world. Even if the estimate of transport across the equator is significantly wrong, the other sub-regions of the NAO are also accumulating N (Michaels et al. this volume). The injection of anthropogenic N to the NAO by cross shelf transport from major rivers and atmospheric deposition is small relative to natural N sources in the NAO. However, if we consider as anthropogenic the possible increase in N-fixation in the Sargasso Sea from increased Saharan dust (75% higher than before the drought) this would add another 2,800 to 4,800  $\text{Gmol yr}^{-1}$  to the anthropogenic flux (see later discussion) (Michaels et al. this volume).

Overall, the greatest areas of uncertainty in the N budget for the NAO&W are (1) storage vs. denitrification losses in the watershed, (2) natural N-fixation in the watershed, (3) N-fixation in the open ocean, and (4) the exchange of N between the NAO and the SAO. It is important to gain additional information on all of these fluxes. Until we learn the balance between N storage vs. denitrification in the NAO watershed we are unable to assess to what degree anthropogenic N is accumulating in continental ecosystems. In addition, until we have better information on natural terrestrial N-fixation we are unable to estimate the rate at which human activities are fixing N relative to natural processes. Our uncertainty about N-fixation rates in the ocean and the degree that the NAO is a N source or a sink relative to the SAO make it difficult to determine not only the natural N cycle for the NAO but also the possible impact of Fe input on N-fixation in the NAO.

### *P distribution patterns in the NAO&W*

The distribution patterns of N and P through the environment are quite different. N has a significant gas phase, and is distributed by atmospheric as well as hydrologic processes. P has no stable gas phase in the atmosphere and is only transported on particles, limiting its atmospheric lifetime and hence its



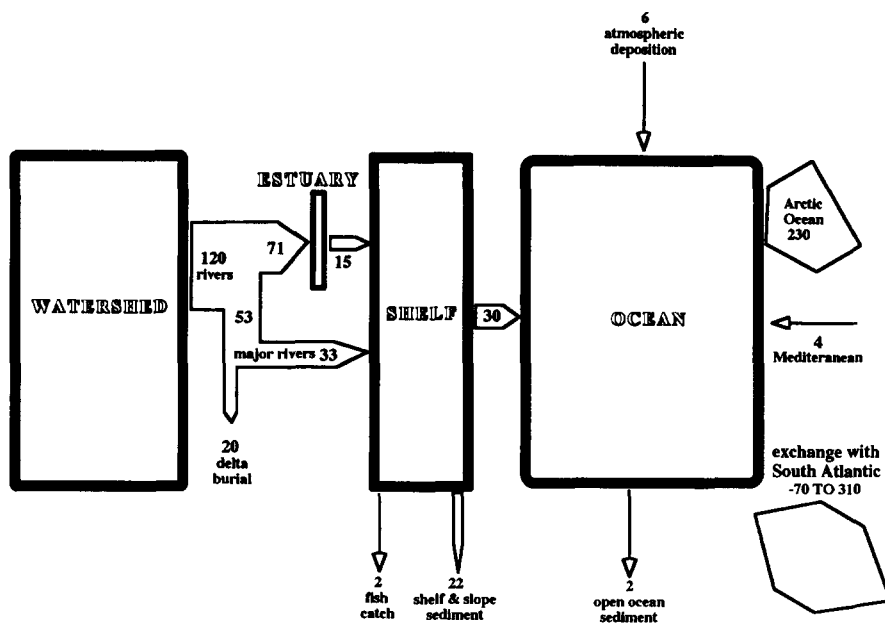


Figure 4. The P budget for the North Atlantic Ocean ( $\text{Gmol yr}^{-1}$ ).

global dispersion. Hence atmospheric and riverine N inputs into the marine environment are both important (Figure 2), while P inputs from rivers are substantially greater than those from the atmosphere (Figure 4). There is another critical difference between N and P. N mobilized by human activities can be “unmobilized” by both long term storage (e.g., sediments) and by denitrification, which returns N to an inactive state ( $\text{N}_2$ ). For P, there is no process equivalent to denitrification. Once mobilized by human activity, it accumulates in either a short term reservoir (biomass) or a long term reservoir (sediments). Given the importance of hydrologic transport for P, and its potential for storage in sediments, we focus our effort on the coastal, shelf and open ocean subsystems of the NAO&W.

Annually, about  $120 \text{ Gmol yr}^{-1}$  P are lost from the watershed via rivers of which about 53  $\text{Gmol P}$  are from large rivers (mostly the Amazon) and the remainder from smaller rivers that flow into estuaries (Figure 4). Most of the riverine P is deposited to delta, estuarine, shelf and slope sediments ( $\sim 100 \text{ Gmol yr}^{-1}$ , Figure 4). Only about  $30 \text{ Gmol yr}^{-1}$  are injected into the open ocean from rivers. Another 160–300  $\text{Gmol yr}^{-1}$  are injected to the NAO from the Arctic Ocean (230  $\text{Gmol yr}^{-1}$ , Figure 4). It appears that the NAO may be accumulating P at a rate of about 0.1 % per year (Michael et al. this volume). However, given the relatively large uncertainties around

many of the estimates of P fluxes and stocks, this small difference may not be separable from a balance.

### *Impact of anthropogenic N and P on the NAO&W*

*Terrestrial systems.* NO<sub>x</sub> emissions from fossil fuel combustion have increased the oxidizing potential of the atmosphere on a regional basis in the watershed of the NAO with measurable impacts on human health. The same emissions have also increased precipitation acidity in wide spread portions of the watershed of the NAO, with concomitant impacts on terrestrial and aquatic ecosystems (e.g., Grennfelt et al. 1995). The addition of fertilizer N to the watershed of the NAO has purposefully increased productivity of agricultural ecosystems, resulting in the fixation of more carbon and other elements associated with plant and animal growth. There are also inadvertent increases in productivity in other ecosystems which result in alterations to other elemental cycles. Persistent atmospheric deposition of fixed N to forest ecosystems may eventually lead to retrogressive changes in ecosystem function. Aber et al. (1989) suggested that forests in the northeastern United States are becoming N sources rather than N sinks because of extensive N deposition resulting in N supply in excess of demand. Not only are the forests potentially impacted by the high rate of atmospheric deposition but they are also N sources to downstream ecosystems and to the atmosphere (N<sub>2</sub>O emissions). The emission and subsequent deposition of N results in increased growth of forests and sequestering of C and other nutrients (Schindler & Bayley 1993; Schimel et al. 1995).

*Coastal systems.* Riverine injection of anthropogenic N and P to coastal ecosystems has the potential to increase productivity (Howarth 1988; Turner & Rabelais 1991; Vitousek & Howarth 1991; NRC 1993; NRC 1994; Nixon 1995; Howarth et al. 1995). The importance of the N versus P contribution to increased productivity depends on their relative injection rates. If the N:P ratio is <16 ("Redfield" ratio), theoretically N is the limiting nutrient; if the ratio is >16, P is the limiting nutrient. The N:P ratios vary by region. In the Amazon, it is 4.5:1; waters flowing into the western Gulf of Mexico have a 271:1 ratio. However, for most of the riverine fluxes to the NAO coastal region, the N:P ratios are within a factor of 2 or 3 of the "Redfield" ratio, suggesting that estuaries in most of these regions have the potential to be either N or P limited, depending on the bioavailability of the N and P, and the estuarine processes that alter the ratio (Howarth et al. this volume).

Anthropogenic N in rivers probably does not directly impact the open ocean. Riverine N is primarily denitrified in the shelf region (except for the Amazon), and thus never reaches the open ocean. However, an indirect

impact is that anthropogenic N in rivers results in a decrease in the open ocean N export to the shelf. In the pre-industrial world, if shelf denitrification was unchanged from today, than supply of N from the open ocean was perhaps about double of what it is today. If this is correct, then the oceanic accumulation may have an additional anthropogenic cause.

*Open ocean.* Anthropogenic N can be deposited to the open ocean, but at rates that are generally small compared to other sources (Michaels et al. 1993). While anthropogenic N probably does not have a major impact on the overall biogeochemistry of the NAO, there may be episodic impacts on productivity in stratified regions of mid-ocean gyre, where upwelling supply of N is limited (Owens et al. 1992; Michaels et al. 1993). In these situations, P limitation may be a confounding factor. The N:P ratio of the deposition of reactive N and P from the atmosphere to the ocean varies systematically across the basin. Reactive N is primarily from pollution sources and is maximal near the U.S. coastline and decreases rapidly to the east. P deposition is primarily with dust and thus has a maximum near the coast of Africa and declines to the west. As a result, the ratio of reactive N to P varies from values of 10–30 near the African coast to values of 500–1500 near the U.S. coastline (calculated from Prospero et al. this volume). This analysis does not include organic N or P; however, since levels of organic phosphorous are low, the N:P ratio is dominated by the substantial pools of organic nitrogen. Given that the N:P ratios are always greater than the Redfield ratio, it is possible that the P deposition may regulate the impact on surface biology. However, there are confounding factors; e.g., much of the deposited P is tightly bound in insoluble compounds such as apatite and, consequently, the P is not readily available to organisms.

Phytoplankton in the surface waters of the Sargasso Sea are exposed to low concentrations of both reactive N and phosphate; in situ concentrations are below the detection limits of standard analysis for both nutrients. Thus, the instantaneous growth rates of most phytoplankton could be co-limited by both nutrients. Some organisms may have the ability to fix atmospheric N and these would obviously be limited by the availability of phosphate. The atmospheric deposition patterns modify this picture. In nearly all cases the deposition ratio of N and P is higher than the traditional stoichiometry of phytoplankton growth, the Redfield ratio of 16N:1P. Thus, a deposition event, even if it adds considerable reactive N to the surface mixed layer, will rarely add enough phosphate to allow complete utilization of the N. Thus, in these deposition events, surface populations are likely phosphate limited. Since the required phosphate supply to sustain the N-fixation postulated by Michaels et al. this volume, is very large compared to the atmospheric

deposition, the phosphate to support N-fixation must come from the stocks in the thermocline. They suggest that the migration behavior of the large and colonial organism responsible for the nitrogen-fixation allow them access to these deeper phosphate stocks. Consequently, the nutrient limitation patterns for the surface community are probably determined by the dynamics of the phosphate transport from below and not the atmospheric deposition.

There may be an indirect impact of human activities on the open ocean N cycle. There is an interesting and potentially very important interaction between the N cycle and Fe. It is proposed that Fe deposition to the NAO by atmospheric transport from Africa is supporting N-fixation rates in the NAO (Prospero et al. this volume; Michaels et al. this volume). A substantial fraction of the mineral dust carried to the NAO appears to be mobilized as a consequence of human activities in Africa compounded by rainfall variability (Prospero et al. this volume); thus, the impact of dust-borne Fe on N-fixation rates in the NAO could be regarded as being largely an anthropogenic effect. Also, because dust transport is highly variable from year-to-year due to changes in weather and climate (Prospero & Nees 1986) the impact of Fe deposition to the ocean could also vary greatly. This linkage of emission processes in Africa to ecosystem productivity in far downwind ecosystems is not new – it has also been proposed for the supply of P to portions of the Amazon forest (Swap et al. 1992). Further, the nitrogen-fixation that occurs in the Sargasso Sea would require an additional uptake of dissolved  $\text{CO}_2$  of approximately  $0.3\text{--}1.2 \text{ Gt yr}^{-1}$  (Michaels, this volume). This carbon is remineralized at depths with residence times of decades, so the process does not result in a long-term sequestration of surface or atmospheric carbon. However, if the rate of nitrogen-fixation is regulated by the highly variable dust deposition, then the resulting variations in N-fixation rate could yield significant interannual fluctuations in the carbon drawdown and perhaps affect the interannual variations in atmospheric  $\text{CO}_2$  as well.

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