

Atmospheric Deposition of Nitrogen: Implications for Nutrient Over-enrichment of Coastal Waters

HANS W. PAERL^{1,*}, ROBIN L. DENNIS^{2,†}, and DAVID R. WHITALL³

¹ *Institute of Marine Sciences, University of North Carolina at Chapel Hill, 3431 Arendell Street, Morehead City, North Carolina 28557*

² *U.S. Environmental Protection Agency, National Oceanic and Atmospheric Administration, Atmospheric Modeling Division, Research Triangle Park, North Carolina 27711*

³ *Department of Civil and Environmental Engineering, Syracuse University, Syracuse, New York 13244-1190*

ABSTRACT: Atmospheric deposition of nitrogen (AD-N) is a significant source of nitrogen enrichment to nitrogen (N)-limited estuarine and coastal waters downwind of anthropogenic emissions. Along the eastern U.S. coast and eastern Gulf of Mexico, AD-N currently accounts for 10% to over 40% of new N loading to estuaries. Extension of the regional acid deposition model (RADM) to coastal shelf waters indicates that 11, 5.6, and 5.6 kg N ha⁻¹ may be deposited on the continental shelf areas of the northeastern U.S. coast, southeast U.S. coast, and eastern Gulf of Mexico, respectively. AD-N approximates or exceeds riverine N inputs in many coastal regions. From a spatial perspective, AD-N is a unique source of N enrichment to estuarine and coastal waters because, for a receiving water body, the airshed may exceed the watershed by 10–20 fold. AD-N may originate far outside of the currently managed watersheds. AD-N may increase in importance as a new N source by affecting waters downstream of the oligohaline and mesohaline estuarine nutrient filters where large amounts of terrestrially-supplied N are assimilated and denitrified. Regionally and globally, N deposition associated with urbanization (NO_x, peroxyacetyl nitrate, or PAN) and agricultural expansion (NH₄⁺ and possibly organic N) has increased in coastal airsheds. Recent growth and intensification of animal (poultry, swine, cattle) operations in the midwest and mid-Atlantic regions have led to increasing amounts of NH₄⁺ emission and deposition, according to a three decadal analysis of the National Acid Deposition Program network. In western Europe, where livestock operations have dominated agricultural production for the better part of this century, NH₄⁺ is the most abundant form of AD-N. AD-N deposition in the U.S. is still dominated by oxides of N (NO_x) emitted from fossil fuel combustion; annual NH₄⁺ deposition is increasing, and in some regions is approaching total NO₃⁻ deposition. In receiving estuarine and coastal waters, phytoplankton community structural and functional changes, associated water quality, and trophic and biogeochemical alterations (i.e., algal blooms, hypoxia, food web, and fisheries habitat disruption) are frequent consequences of N-driven eutrophication. Increases in and changing proportions of various new N sources regulate phytoplankton competitive interactions, dominance, and successional patterns. These quantitative and qualitative aspects of AD-N and other atmospheric nutrient sources (e.g., iron) may promote biotic changes now apparent in estuarine and coastal waters, including the proliferation of harmful algal blooms, with cascading impacts on water quality and fisheries.

Introduction

Human population growth and urban, industrial, and agricultural expansion have profoundly impacted the quantities and composition of nitrogen (N)-containing pollutants released to the environment (Vitousek et al. 1997). In particular, human encroachment on the coastal zone has been accompanied by a precipitous rise in land-based N nutrients released as either point sources (i.e., wastewater, industrial discharges, stormwater overflow discharges) or diffuse nonpoint agricultural, urban, and rural discharge. A strong and direct relationship between human expansion and accel-

erating riverine N inputs to coastal waters testifies to this impact (Peierls et al. 1991; Howarth et al. 1996). Diverse studies in a broad range of estuarine and coastal waters receiving anthropogenic N enrichment have shown these waters to be N-limited; namely, N supply controls the rate of primary production (Dugdale 1967; Ryther and Dunstan 1971; D'Elia et al. 1986; Nixon 1986, 1995). It follows that N enrichment has been linked to accelerated primary production, or eutrophication, in these waters (Nixon 1986, 1995). N-enhanced eutrophication may be manifested as increased frequencies, magnitudes, and persistence of suspended algal (phytoplankton) blooms (Paerl 1988; Richardson 1997). Phytoplankton blooms may be toxic and accumulate as large masses of organic matter in the sediments, fueling excessive oxygen consumption in the form of hypoxia (2–3 mg l⁻¹

* Corresponding author; e mail: hans.paerl@unc.edu.

† On assignment to the National Exposure Research Laboratory, U.S. Environmental Protection Agency, Research Triangle Park, North Carolina 27711.

O₂) and anoxia (no detectable O₂), conditions that are stressful or fatal to resident finfish and shellfish communities (Winn and Knott 1992; Justic et al. 1993; Diaz and Rosenberg 1995; Paerl et al. 1998; Rabalais et al. 1999).

Management of coastal eutrophication has focused on identifying and reducing excessive N inputs to these N-sensitive waters (Environmental Protection Agency 1989; National Research Council 2000). Both point and nonpoint land-based sources have been identified as targets for N input controls and a variety of land-based nutrient reduction and mitigation strategies have evolved to tackle the problem. When identifying key external or new N sources responsible for over-enrichment of N sensitive waters, it has become evident that land-based sources are not the only culprits. Among anthropogenic sources of new N to estuarine and coastal waters, atmospheric deposition of N (AD-N) is now recognized as highly significant and increasingly important (Paerl 1985, 1995; Fisher and Oppenheimer 1991; Chesapeake Bay Program 1994; Valigura et al. 1996; Jaworski et al. 1997; Ecological Society of America 1998; Environmental Protection Agency 1999). In the past century alone, anthropogenically-mediated atmospheric N emissions have increased by nearly 10-fold (Howarth 1998). It is estimated that from 10% to over 40% of new N supplied to these waters is of atmospheric origin (Valigura et al. 1996, 2000). In some regions (e.g., mid-Atlantic, Northeast, Tampa Bay), the atmosphere is a dominant source of new N (Valigura et al. 1996). This underscores the need to better quantify AD-N inputs on local and regional scales, and to understand ecosystem-level biogeochemical, trophic, water quality, and habitat responses to this growing N source. AD-N is also an emerging new N source in developing nations experiencing urban, agricultural, and industrial expansion in coastal regions.

Because of its large scale and magnitude, AD-N plays a central role in coastal N budgets, biogeochemical, trophodynamic, water quality, and habitat changes in downwind waters. We must clarify the spatiotemporal linkage of AD-N emissions, transport, and fate on local, regional, and larger (ocean basins, global) scales; the absolute and relative (to land-based sources) roles AD-N plays as a new N source; the sphere of influence of AD-N as a new N source in N sensitive estuaries and coastal regions; the chemical constituents in AD-N; and the biological availability and ecological roles of these AD-N constituents, as they pertain to mechanisms of eutrophication and algal and higher plant composition, including harmful algal bloom species.

We examine and synthesize information pertain-

to the roles of AD-N in coastal nutrient over-enrichment, biogeochemical alterations, and trophic change. We attempt to integrate knowledge of the sources, composition, deposition and ecological responses over scales ranging from ecosystem to regional levels, and address requirements and options for managing this growing and evolving source of N in coastal waters.

Atmospheric N Composition and Sources

Atmospherically-deposited N may be either wet in origin (if N is dissolved in precipitation such as rain or snow), or dry (the settling of N-containing particles and gases to land and water surfaces). AD-N deposited in wet or dry forms may be deposited directly, or it may take an indirect route if it enters the estuary via runoff and groundwater from the surrounding watershed. The majority of atmospheric N exists as inorganic N in two principal forms: oxidized and reduced.

Oxidized N is found in the form of gaseous nitric acid (HNO₃), nitrogen dioxide (NO₂), and aerosol nitrate (NO₃). Contributions from nitrogen dioxide are minimal and will not be considered further. Nitric acid is the parent form of oxidized N. It is a termination product of the oxidation of nitrogen oxide (NO_x = NO + NO₂) and volatile organic compounds as part of atmospheric photochemistry (Environmental Protection Agency 1996; Seinfeld and Pandis 1998). NO₃ exists as ammonium nitrate; it results from the partitioning of a fraction of nitric acid into particulate form in the presence of ammonia (NH₃; Seinfeld and Pandis 1998). The principal source of nitric acid and NO₃ in North America is the combustion of fossil fuels (e.g., by coal and oil-fired power plants, automobiles, and other forms of transportation) that produces precursor emissions of NO_x, primarily NO.

Reduced N in the atmosphere exists in the form of gaseous NH₃ and aerosol ammonium (NH₄). NH₄ results from chemical reactions involving sulfuric acid droplets, nitric acid, and NH₃ that partitions a fraction of NH₃ into fine particulates: ammonium sulfate, ammonium bisulfate, ammonium nitrate, and other complex aerosol mixtures (Seinfeld and Pandis 1998). The majority of NH₃ is directly emitted into the atmosphere from agricultural sources such as fertilizer application and confined animal operations.

Organic nitrogen (ON) is also an important fraction of AD-N (Correll and Ford 1982; Timperley et al. 1985; Mopper and Zika 1987). The sources and composition of atmospheric ON are not well understood, although rainfall in agricultural areas tends to be relatively enriched in dissolved organic nitrogen (DON; e.g., Timperley et al. 1985). DON

accounts for ~15% to 30% of wet N deposition in coastal and open ocean waters (Correll and Ford 1982; Cornell et al. 1995; Peierls and Paerl 1997; Russell et al. 1998).

Atmospheric N Deposition Processes

All forms of inorganic N are very water soluble and not influenced significantly by aqueous chemistry so all forms wet deposit equally well. Dry deposition is significantly influenced by the form of the inorganic N and the physical characteristics of the surface to which it is deposited. Dry deposition flux depends on both the ambient concentration and the chemical's affinity for deposition (deposition velocity). Gaseous nitric acid deposition velocities exceed those of NH_3 , but at moderate to high concentrations of NH_3 they are the same order of magnitude (Duyzer 1994; Fowler et al. 1998; Meyers et al. 1998). At low concentrations of NH_3 there is a compensation point and plants can become a source of NH_3 rather than a sink (Langford and Fehsenfeld 1992; Duyzer 1994; Fowler et al. 1998; Wyers and Erisman 1998). Over land, gaseous NH_3 and nitric acid are very sticky (high deposition velocity), whereas, the small ($< 0.5 \mu\text{m}$) inorganic N particles are much less likely to deposit (low deposition velocity). The gaseous deposition velocity is an order of magnitude higher than the small particulate deposition velocity. For both gases and aerosols, the deposition velocity to open, freshwater surfaces is roughly 2 to 5 times lower than to terrestrial surfaces. Over salt water, sea salt aerosol will preferentially and efficiently partition interstitial nitric acid gas to NO_3 aerosol associated with supermicron particles (1–5 μm and greater; Keene and Savoie 1998). This shift in aerosol size from 0.1–0.5 μm over land, a size not affected by gravity, to the order of 2–5 μm over salt water, a size significantly affected by gravity, will increase the particle NO_3 deposition velocity by more than an order of magnitude (Slinn 1982; Davidson and Wu 1990; Torseth and Semb 1998; Pryor et al. 1999). Over open salt water, particle NO_3 deposition will dominate dry deposition and it can be the same order of magnitude as NO_3 wet deposition. The thermodynamic and phase partitioning relationships do not favor NH_3 partitioning to supermicron particles and it tends to be found principally on the fine particles $< 0.5 \mu\text{m}$.

Due to the large difference in gaseous and fine particulate deposition velocities, dry deposition rates of oxidized and reduced N to land are different, which affect ratios of wet to dry deposition for the two types of inorganic N. Modeling analysis with the Extended Regional Acid Deposition Model (Extended RADM) indicates that the partitioning of ambient concentrations of nitric acid and

NH_3 into the fine particulate species of nitrate and ammonium is basically reversed with respect to one another for the land areas of coastal states of the eastern U.S. (Mathur and Dennis 2000). The fractions of $\text{NO}_3/(\text{NO}_3 + \text{HNO}_3)$ and $\text{NH}_4/(\text{NH}_4 + \text{NH}_3)$ have medians of 0.23 and 0.74, respectively, over the eastern and Gulf Coast estuaries. This same pattern is observed in northern Europe (Sorteberg et al. 1998) and in CASTNet data for oxidized N in the eastern U.S. (Clarke et al. 1997). The dry deposition of oxidized N will be significantly larger than that of reduced N, whereas wet deposition is hardly or not at all influenced by the differences in partitioning. Model analyses suggest that wet and dry deposition to terrestrial surfaces of oxidized forms of N will be roughly comparable on a long-term annual basis. Clarke et al. (1997) suggest that wet and dry deposition of reduced forms of N to land surfaces will be unequal, with wet deposition being roughly a factor of 2 to 3 larger on an annual basis. Over salt water, wet and dry deposition of oxidized forms of N are also expected to be comparable on an annual basis. For reduced forms of N, wet and dry deposition over salt water will be unequal, with wet deposition being much larger. Over estuaries with a fresh-to-salt water gradient at the surface, there will be a gradient in the dry deposition of oxidized N, tracking the increase with increasing salinity of nitrate attached to supermicron-sized particles.

Because dry deposition velocities for open freshwater are lower than those for land, the delivery of direct AD-N to estuaries is expected to be dominated by wet deposition, even on an annual basis. National Acid Deposition Program (NADP) data show that while more oxidized N than reduced N was being deposited in the 1980s, the reduced fraction is increasing with time (Fig. 1). About the same mass of oxidized and reduced N is currently being deposited to coastal areas. Wet deposition is an acute or episodic form of AD-N, while dry deposition is a chronic, low-level source of AD-N. If NH_3 emissions sources are large and nearby, as has been shown for hog operations in North Carolina (Aneja et al. 2000; Whitall and Paerl 2001) then the dry deposition of reduced N may be much larger than indicated here, possibly comparable to the wet deposition. The exact relationship between wet and dry reduced and oxidized N needs to be locally established for individual airsheds and watersheds with the help of both measurements and modeling. The terrestrial release of atmospherically deposited N to the rivers is most likely to be in the form of oxidized N (Stoddard 1993), so the major source of atmospherically deposited NH_3 is likely to be direct deposition to the estuary surface.

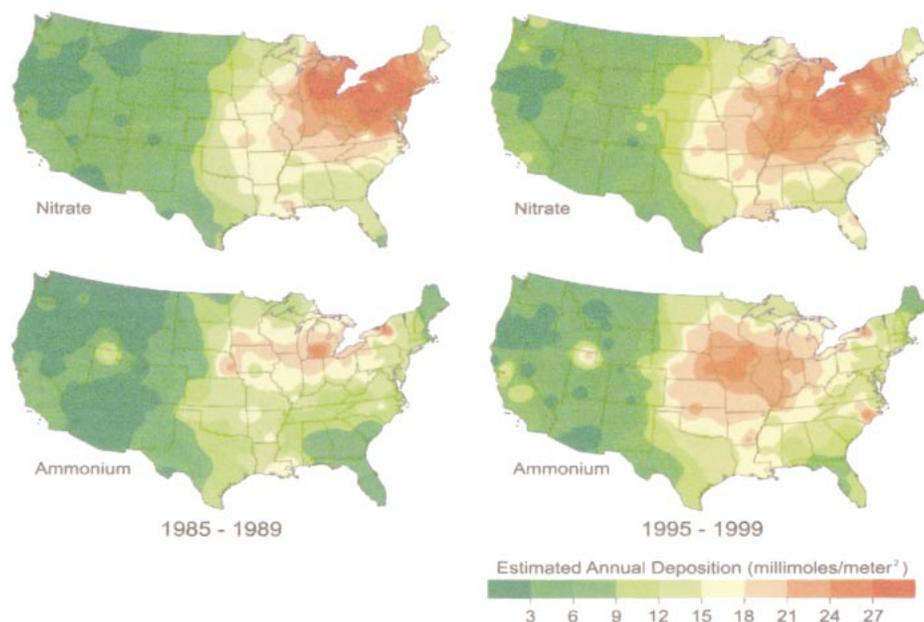


Fig. 1. Estimated annual wet deposition of nitrate and ammonium for the mainland U.S. during the intervals 1985–1989 and 1995–1999. Annual nitrogen deposition is given as mmol m^{-2} . Data and figure are from the National Acid Deposition Program (NADP) network of collection sites, 1999 summary (NADP 2000).

The N Airshed for Estuaries

To address AD-N contributions to the estuaries, it is necessary to establish a sense of the sources of the AD-N. This can be accomplished through the concept of an airshed. The atmosphere does not have a lateral boundary and the flow of atmospheric chemicals to an estuary does not have a well-defined beginning as does the flow of surface waters. The boundary can be defined in terms of the relative climatological range of the majority of the deposition from a source and in terms of deposition efficiency. The concept of a normalized range of influence of a source region has been quantified to produce a prescription for constructing principal airsheds for watersheds (Dennis 1997; Dennis and Mathur unpublished data). The distance at which the accumulated deposition from a source region encompasses $1/e$ of its total continental deposition, roughly the 65% contour of accumulation, is taken as the source's normalized range of influence. The RADM (Chang et al. 1987) and the Extended RADM (Mathur and Dennis 2000) have been used to define the range of influence of over 100 source regions across the eastern U.S. The normalized range of influence of annual oxidized-N deposition from NO_x emissions for the eastern U.S. is approximately 400–600 km in the prevailing direction of transport. The distance is shorter in a direction that opposes the prevailing wind. The normalized range of influence of annual reduced-N deposition from NH_3 emissions for the eastern

U.S. is approximately 75% of the range for oxidized N or 300–450 km. The modeling approach used to develop annual averages (Dennis et al. 1990; Brook et al. 1995a,b) gives a climatological average representative of at least 10 years of meteorology, not a single specific year. These ranges are consistent with a residence time for N compounds in the lower atmosphere, controlled by deposition loss processes, of approximately 1 to 2 d for oxidized N and about 1 d for reduced N. The atmospheric lifetime measured by deposition will be slightly shorter than the typical lifetime of the chemicals in the atmosphere, because the lifetimes in the upper troposphere, which are longer, are being ignored. Precipitation scavenging plays an important role in determining the deposition lifetimes for the eastern U.S.

Interpretation of model evaluation results (e.g., Cohn and Dennis 1994) suggests that most likely the size of the oxidized-N range of influence is biased towards short distances. Overprediction of wet deposition by 20% on average for both oxidized N and reduced N is a defining evaluation result. Evaluation results for reduced N are mixed. Errors suggest biases in both directions that potentially offset each other. The range of influence for reduced N appears to be a reasonable best estimate. In modeling studies for northern Europe the range for oxidized N is somewhat longer than for the eastern U.S., and the difference in the transport range between oxidized N and reduced N is larger (Hov et

TABLE 1. Physical-chemical characteristics of major U.S. Atlantic and Gulf of Mexico Airsheds. (R. Dennis, USEPA, Research Triangle Park, North Carolina).

Watershed	Size (km ²)	Size Factor Over Watershed Area	% Oxidized-N Deposition Explained	Airshed NO _x Emissions as % of Eastern North America	Efficiency of Deposition (% Deposition per % Emission)
Casco Bay	624,000	214	47	10	4.7
Great Bay	547,200	214	60	13	4.6
Narragansett Bay	595,200	138	73	18	4.1
Long Island Sound	905,600	22	70	23	3.0
Hudson/Raritan Bay	912,000	22	62	25	2.5
Barnegat Bay	505,600	361	67	16	4.2
Delaware Bay	729,600	22	75	26	2.9
Delaware Inland Bays	326,400	584	52	12	4.3
Chesapeake Bay	1,081,600	6.5	76	34	2.2
Pamlico Sound	665,600	25	63	18	3.5
Winyah Bay	886,400	19	69	24	2.9
Charleston Harbor	806,400	20	56	18	3.1
St. Helena Sound	588,800	48	59	11	5.4
Altamaha	678,400	18	68	13	5.2
Tampa Bay	256,000	45	76	5	15.2
Apalachee Bay	441,600	31	50	9	5.6
Apalachicola Bay	812,800	16	69	17	4.1
Mobile Bay	992,000	8.7	68	17	4.0
Lake Pontchartrain	659,200	17	63	11	5.7
Barataria-Terrebonne	409,600	55	63	8	7.9

al. 1994; Hov and Hjøllø 1994). The higher NH₃ emissions in Europe and the drier north, together with the judgments about the effect of biases on oxidized-N ranges, are the most likely explanations for the differences. Hence, the differences in transport range reflect differences in emissions and climatology between the two continents. Oxidized-N principal airsheds have been defined and characteristics developed for 20 watersheds along the U.S. East Coast and Gulf Coast. Oxidized-N airshed characteristics are given in Table 1 for all of the watersheds and are summarized in Table 2.

The airsheds are large compared to watersheds,

even though only the area over the continent is counted. Example airsheds for the East Coast and Gulf Coast are shown in Fig. 2. Most of the airsheds are from 15–200 times larger than the drainage area of the watersheds receiving the N deposition. The variation in size representative of the distribution of airsheds, defined by the span between the 20th and 80th percentiles of the rank-ordered sizes, is about a factor of two, from 475,000 to 900,000 km² (Table 2). The size of the airshed is influenced by variations in the climatological range across the eastern U.S. and the size of the watershed, but the variation in airshed size is much

TABLE 2. Summarized characteristics of principal airsheds for oxidized-nitrogen deposition.

Percentiles	Principal Oxidized-N Airshed Area (km ²)	Size Factor over Watershed Area	% Oxidized-N Deposition Explained by Airshed Emissions	Airshed NO _x Emissions as % of Eastern North American Emissions
Maximum	1,081,600 Chesapeake Bay	584 Delaware Inland Bays	76 Chesapeake Bay, Tampa Bay	34 Chesapeake Bay
80th	896,000 Long Island Sound, Winyah Bay	176 Great Bay, Narragansett Bay	71.5 Narragansett Bay, Long Island Sound	23.5 Winyah Bay, Long Island Sound
50th	662,000 Pamlico Sound, Lake Pontchartrain	23.5 Pamlico Sound, Long Island Sound	65 Barnegat Bay, Pamlico Sound	16.5 Apalachicola Bay, Barnegat Bay
20th	474,000 Barnegat Bay, Apalachee Bay	17.5 Altamaha Lake Pontchartrain	57.5 St. Helena Sound, Charleston Harbor	10.5 St. Helena Sound, Casco Bay
Minimum	256,000 Tampa Bay	6.5 Chesapeake Bay	47 Casco Bay	5 Tampa Bay
80th/20th	1.9	10.1		
2 (80th–20th) spread as % of 50th	± 32%	na	± 11%	± 39%

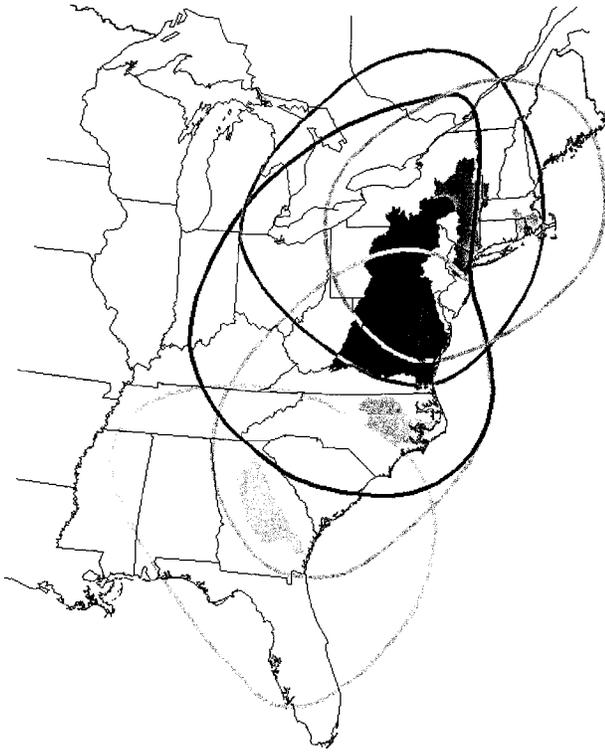


Fig. 2. Oxidized-nitrogen principal airsheds for the U.S. east coast: Narragansett Bay, Hudson River/Raritan Bay, Chesapeake Bay, Pamlico Sound, and Altamaha Sound.

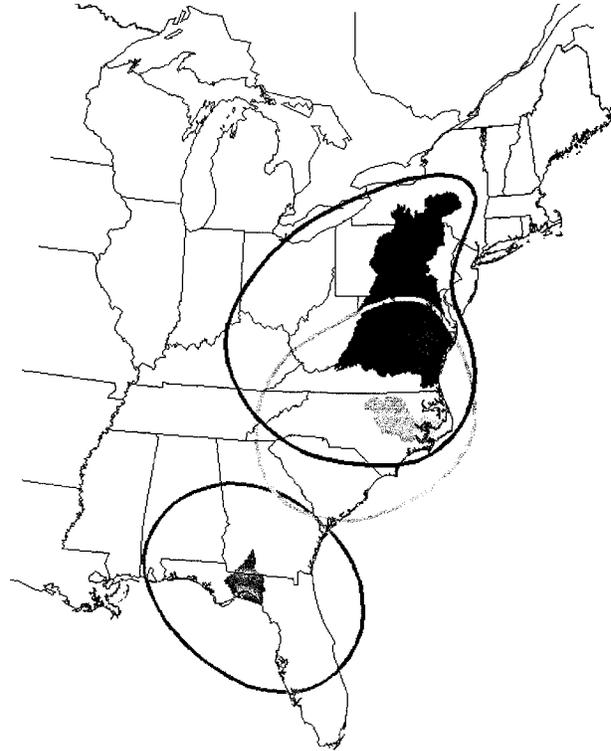


Fig. 3. Reduced-nitrogen principal airsheds for Chesapeake Bay, Pamlico Sound, and Apalachee Bay.

smaller than that for watershed area because the atmospheric range of influence is large. The airshed for Tampa Bay is unusually small because of its location relative to the geography of Florida. The NO_x emissions from within the airshed boundary explain from 47% to 76% of the oxidized-N deposition to the watershed drainage area. For slightly more than half of the watersheds, the emissions from the airshed explain between 60% and 70% of the deposition. When the percent explained is 70% or higher, emissions within or close to the watershed are important. When the percent explained is $\sim 55\%$ or lower, long-range transport of the oxidized N from outside the airshed to the watershed is very important.

Characteristics of reduced-N airsheds have been developed for three watersheds along the East and Gulf Coasts (Table 3 and Fig. 3). The reduced-N

airsheds are smaller than those for the oxidized N, although they are still much larger than the watersheds. The two reduced-N airsheds in the mid-Atlantic region are 61% and 64% as large as the oxidized-N airsheds, whereas, the Gulf Coast reduced-N airshed is 71% as large as the oxidized-N airshed. In the Southeast, where there is more rainfall, the two airsheds appear closer in size. It is surprising that the percent of the reduced-N deposition explained by the NH_3 emissions from the airshed is smaller than for oxidized N, even though the range of influence for reduced N is shorter. The percent explained for reduced N ranges from 49–60%. The fact that as much of the reduced-N deposition is accounted for by a much smaller fraction of the eastern North American NH_3 emissions compared to the fraction for the NO_x emissions indicates that local deposition is indeed important. Transport is three-dimensional, and because most

TABLE 3. Characteristics of principal airsheds for reduced-nitrogen deposition.

Watershed	Principal Reduced-N Airshed Area (km ²)	Reduced-N Area as % of Oxidized-N Area	% Reduced-N Deposition Explained by Airshed Emissions	Airshed NH_3 Emissions as % of Eastern North American Emissions
Chesapeake Bay	688,000	64%	55%	11%
Pamlico Sound	406,400	61%	60%	6.8%
Apalachee Bay	313,600	71%	49%	4.3%

of the reduced-N deposition is in the form of wet deposition, very long-range transport of small, diffuse concentrations collectively can have an important depositional impact. Emissions in the central part of the continent are generally high, adding to the importance and size of a diffuse background of reduced N entering coastal regions.

The principal airsheds provide perspective regarding the geographic source of the majority of the inorganic N deposition to the estuaries. The emissions from within the airsheds explain a clear majority of the oxidized-N deposition and at least half of the reduced-N deposition to the watersheds. Long-range transport to the watersheds of inorganic N from hundreds of km away is clearly important for both forms of AD-N. Long-range transport is especially important for reduced N, a conclusion somewhat contrary to conventional wisdom. Because modeling sensitivity studies are consistent with observed trends in North Carolina (Dennis and Mathur unpublished data), conclusions based on these modeling results seem reasonable.

Biogeochemical, Trophic, and Water Quality Impacts of AD-N on Estuarine and Coastal Ecosystems

Sensitivity and response of a receiving estuary to specific N inputs are dependent on interacting hydrologic, morphologic, and biogeochemical characteristics. Surface area, volume, and depth, combined with hydrologic throughput (flushing), determine water residence time, a key factor controlling nutrient concentrations, loading rates, and biological response to N inputs. The estuary's response varies along its longitudinal salinity gradient, which strongly affects plant and animal community composition and activity. In an estuary's predominantly fresh headwaters, riverine (terrigenous) discharge dominates new N inputs. Further downstream, in mesohaline segments of the estuary, significant fractions of the terrigenous N load are assimilated by phytoplankton and benthic flora, or microbially denitrified to biologically unreactive N₂ gas (Kennedy 1983; Nixon 1986; Seitzinger 1988; Boynton et al. 1995). Inputs of N that result from direct deposition, however, can bypass this estuarine N filter (Kennedy 1983; Paerl 1995, 1997), so that AD-N assumes an increasingly important role as a new source of N in lower estuarine, sound, and coastal waters below the biological N filtering zone (Paerl and Whitall 1999). Because of a scarcity of direct measurements, there is a great deal of uncertainty as to how the relative importance of AD-N increases beyond this zone. We must rely on modeling efforts based on meteorology and the known behavior of AD-N constituents.

The Role of AD-N in Estuarine and Coastal N Budgets

There are efforts underway to quantify wet and dry deposition at nearshore and offshore locations worldwide (Duce 1986, 1991; GESAMP 1989; Ecological Society of America 1998). These efforts address estuarine and coastal systems along geographic and trophic gradients. In the U.S., gradients range from the highly impacted northeast and mid-Atlantic to less impacted Gulf of Mexico and Pacific coastal regions (Castro et al. 2000). Included are systems varying in size, water residence time, and trophic state, enabling researchers and managers to make comparisons of the relative importance of AD-N in N budgets, water quality, and trophodynamics. The relative importance of AD-N as a new N source in select estuarine and nearshore coastal systems varies from ~5% in waters most heavily impacted by terrigenous (relative to atmospheric) N inputs (e.g., Mississippi plume region of the northern Gulf of Mexico), to over 30% in waters heavily dominated by AD-N inputs (e.g., Baltic, western Mediterranean, mid-Atlantic and northeast U.S.-Canadian Atlantic coastal regions). A summary of representative N-sensitive waters for which adequate data are available is provided in Table 4.

The Extended RADM was used to develop estimates of the flux of total (wet + dry) oxidized plus reduced N from the atmosphere to the coastal ocean over the continental shelf. The model does not include sea salt, so its influence was incorporated in a post-processing step. Assuming recent estimates of the mean diameter of sea salt aerosol of 2–5 μm for the East Coast of the U.S. are correct and apply to the Gulf Coast as well, then a reasonable assumption is that wet and dry deposition of oxidized N will be approximately equal. Dry deposition from the Extended RADM was adjusted accordingly. The results indicate that sea salt influence is clearly important to the overall flux. Deposition rates of 11, 5.6, and 5.6 kg N ha⁻¹ were calculated for the continental shelf areas of the northeastern U.S. coast (NE), southeast U.S. coast (SE), and eastern Gulf of Mexico (East Gulf), respectively. The average rate was computed for the shelf area out to a 200 m depth isopleth. Several known model biases are expected to roughly cancel out, but there is uncertainty. These rates are considered best estimates with a likely uncertainty of $\pm 30\%$. They are in the same range, but overall somewhat higher than those of Prospero et al. (1996), being 60% higher for the NE and 25% higher for the SE and the East Gulf. For the area covering the continental shelf from 0–200 m depth, the best estimate for annual total N flux

TABLE 4. Estimated contributions of atmospheric deposition of nitrogen (AD-N) to new nitrogen inputs in diverse estuarine, coastal, and open ocean waters. The sources, wet (W) and/or dry deposition (D), and chemical forms, inorganic (I) and/or organic (O), of AD-N are indicated (table adapted from Paerl 1997).

Receiving Waters	% of new N as AD-N	References
Baltic Sea (Proper)	~30 W+D, I	Elmgren 1989; Ambio 1990
Kiel Bight (Baltic)	40% W, I	Prado-Fiedler 1990
North Sea (Coastal)	20–40% W+D, I	GESAMP 1989
Western Mediterranean Sea	10–60% W, I	Martin et al. 1989
Waquoit Bay, Massachusetts	29% W, I+O	Valiela et al. 1992
Narragansett Bay	12% W, I+O	Nixon 1995
Long Island Sound	20% W, I+O	Long Island Sound Study 1996
New York Bight	38% W, I+O	Valigura et al. 1996
Barnegat Bay	40% W, I+O	Moser 1997
Chesapeake Bay	27% W, I+O	Chesapeake Bay Program 1994
Rhode River, Maryland	40% W, I+O	Correll and Ford 1982
Neuse River estuary, North Carolina	34% W, I+O	Whitall 2000
Pamlico Sound, North Carolina	~40% W+D, I	Paerl and Fogel 1994
Sarasota/Tampa Bay, Florida	30% W+D, I	Tampa Bay National Estuary Program 1996
Mississippi River Plume	2–5%, W+D, I+O	Goolsby et al. 1999

from atmospheric deposition of inorganic N to the coastal ocean is 20×10^9 mol N, 5.5×10^9 mol N, and 10×10^9 mol N for the NE, SE, and East Gulf, respectively. The uncertainty of $\pm 30\%$ applies to these estimates as well. These annual fluxes are similar to but somewhat larger than those given in Nixon et al. (1996). The flux from the atmosphere is comparable to the flux from the land (estuaries) to the continental shelf given in Nixon et al. (1996) for the NE and it is about half the flux from the land for the SE and East Gulf areas, although it is difficult to extract a precise estimate for the East Gulf from Nixon et al. (1996). The atmosphere is estimated to contribute a sizable flux of

new N to the U.S. continental shelf on the Atlantic side.

The Neuse River Estuary/Pamlico Sound, North Carolina: A Case Study of the Role of AD-N as an Estuarine New N Source

The Neuse River estuary and Pamlico Sound, located in eastern North Carolina, U.S. (Fig. 4) are representative of shallow N-sensitive estuarine and coastal waters under the influence of growing N inputs and accelerating eutrophication (Copeland and Gray 1991; Bricker et al. 1999). The Pamlico Sound system and its subestuaries are the second largest estuarine complex in the U.S. (5,300 km²) and represent a major fisheries nursery supporting approximately 80% of the mid-Atlantic coastal commercial and sports fisheries (Copeland and Gray 1991). Efforts are under way to examine the role of AD-N in the nutrient budgets and eutrophication dynamics of these waters.

The Neuse and other subestuaries of Pamlico Sound exhibit widespread and chronic N limitation (Hobbie and Smith 1975; Kuenzler et al. 1979; Paerl 1983; Copeland and Gray 1991; Rudek et al. 1991). Anthropogenic N enrichment to these estuaries has been closely linked to eutrophication, algal blooms, hypoxia/anoxia and associated declines in water quality and fisheries resources (Christian et al. 1991; Copeland and Gray 1991; Paerl et al. 1995, 1998). The Neuse estuary receives N inputs from a mosaic of upstream and upwind agricultural, urban, and industrial sources. Fossil fuel combustion and agricultural and industrial N emissions represent a significant and growing source of new N to this system (Paerl and Fogel 1994), reflecting national and worldwide trends (Duce 1986; Luke and Dickerson 1987; Asman 1994; Paerl 1995; Holland et al. 1999). Depending

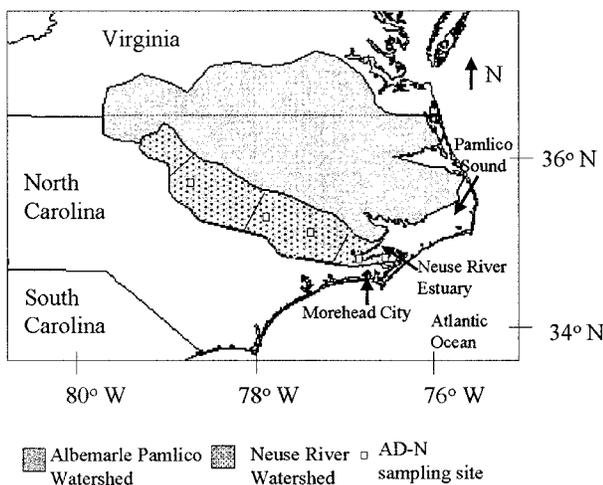


Fig. 4. Location of the Neuse River estuary and Pamlico Sound, eastern North Carolina. The watershed boundaries for both systems are shown, as are the atmospheric deposition collection sites located in watershed zones used to estimate annual atmospheric N deposition inputs to the Neuse River estuary (values given in Table 6). Neuse River watershed zones for the in-stream degradation model (Table 6) are also shown.

TABLE 5. Nitrogen retention model based on GIS land cover data for the Neuse River Watershed. N retention values are from Tyler (1988), Hinga et al. (1991), Fisher and Oppenheimer (1991), and Valigura et al. (1996). The best estimate value is bounded by the highest and median reported retention values (lowest reported values produced unrealistically high AD-N fluxes to estuary; data from Whitall and Paerl 2001).

Model	Percent N Retained by Land				
	Forest	Crop	Pasture	Urban	Other
Highest	100	99.97	99.96	95.3	75
Median	90	80	85	60	60
Best estimate	99	96	97	70	75

on the relationship between watershed-estuary surface areas, degree of watershed N retention, and proximity of atmospheric sources, an important fraction of AD-N is directly deposited on the estuary (Paerl et al. 1995).

The Neuse River estuary has been the focus of a 3-yr (1996–1999) study comparing the terrigenous point and nonpoint sources with wet AD-N flux measurements (NH_4^+ , NO_3^- , and organics) from six sampling locations in the watershed. This AD-N study used a N retention model (Table 5) and an in-stream degradation model (Table 6) to estimate the amount of indirect N deposition reaching the estuary. This value, combined with measured direct deposition fluxes, allowed for the comparison of the total (indirect + direct) AD-N flux to the total new N flux to the estuary. This study has shown that wet AD-N contributes between 15% and 32% of the new or external N flux to the estuary on an annual basis (Table 7), with direct deposition to the estuary alone accounting for 5% of the total new N flux. Deposition is fairly evenly distributed between NH_4^+ , NO_3^- , and organic N. This AD-N flux varies seasonally with the highest fluxes occurring during the summer months, which may be driven by seasonal changes in emissions (Whitall 2000; Whitall and Paerl 2001).

The Neuse River estuary supplies approximately 30% of the Pamlico Sound's freshwater inflow. Downstream of the Neuse's mesohaline N stripping zone, AD-N may represent as much as 40%

of new N inputs to the open Pamlico Sound (Paerl and Fogel 1994; Paerl et al. 1995; Whitall and Paerl 2001).

In the context of ecosystem function, AD-N inputs must be integrated into the overall scheme of N cycling and resultant productivity/eutrophication responses (Fig. 5). This means that we must consider impacts of AD-N contemporaneously and contiguously with internal N cycling (i.e., sediment-water column exchange and regeneration of N), which is known to play a significant role in controlling N availability, productivity, and resultant trophic state of this and other shallow estuaries (Neilson and Cronin 1981; Nixon 1981; Christian et al. 1991; Rizzo et al. 1992).

AD-N and Phytoplankton Production Dynamics

Atmospheric wet and dry deposition introduce into estuaries a variety of biologically-available inorganic compounds (NO_3^- , NH_4^+ , DIN), most of which result from human activities (Likens et al. 1974; Galloway et al. 1994). In addition, ON comprises an additionally significant fraction (from 15% to over 30%) of atmospheric deposition (wet and dry) in coastal watersheds (Correll and Ford 1982; Skudlark and Church 1993; Cornell et al. 1995; Peierls and Paerl 1997; Whitall and Paerl 2001). Although the sources and composition of atmospheric ON are poorly known, recent work (Peierls and Paerl 1997; Seitzinger and Sanders 1999) indicates AD-ON constituents are biologically used and should be included in eutrophication assessments.

Both phytoplankton productivity and community composition respond to chemically diverse AD-N sources. Various bioassay and field surveys show that enrichment with the major atmospheric N constituents NH_4^+ and NO_3^- at natural dilutions results in enhanced phytoplankton primary production and increased biomass (Paerl 1985, 1995; Willey and Paerl 1993; Paerl and Fogel 1994). NO_3^- and NH_4^+ uptake rates vary spatially and seasonally in the Neuse River, suggesting differential community responses to varying N sources (Boyer et al. 1994). Studies on North Sea phytoplankton communities demonstrated differential responses

TABLE 6. In-stream degradation model. Model assumes that the degree to which an N parcel is degraded due to denitrification and settling is directly proportional to travel distance in stream. Model developed by the North Carolina Department of Environment and Natural Resources, Division of Water Quality. Refer to Fig. 4 for the locations of zones 1 through 4 (data from Whitall and Paerl 2001).

Region	Percent N Reaching Estuary
Zone 1 (Above Falls Lake Dam)	10
Zone 2 (Falls Lake Dam to Contentnea Creek)	50
Zone 3 (Contentnea Creek to Streets Ferry)	70
Zone 4 (Below Streets Ferry)	100

TABLE 7. The contribution of AD-N to the total new nitrogen flux to the Neuse River estuary. Columns 1, 5, and 6 are measured values and columns 3 and 7 are derived from model outputs. Total new flux to the estuary (column 6) is equal to the riverine flux measured at the head of the estuary (which inherently includes indirect AD-N), plus point and nonpoint sources below the head of the estuary and plus direct AD-N to the estuary (data from Whittall and Paerl 2001).

Indirect Wet AD-N Deposition (Mg yr ⁻¹)	Model Used	Indirect AD-N Exported to Rivers (Mg yr ⁻¹)	Indirect AD-N Reaching Estuary (Mg yr ⁻¹)	Direct AD-N to Estuary (Mg yr ⁻¹)	Total New N Flux to Estuary (Mg yr ⁻¹)	AD-N as % of Total New N Flux
15,026	Highest	863	718	385	7,408	15
15,026	Median	2,722	1,953	385	7,408	32
15,026	Best Estimate	1,861	1,412	385	7,408	24

to various N sources; providing a mechanism for the structuring of these communities (Stolte et al. 1994). Differential N uptake and growth response to NH_4^+ versus NO_3^- have been attributed to contrasting energy requirements for the assimilation of these compounds (Eppley et al. 1969; Turpin 1991). Under light-limited conditions encountered in turbid estuaries (Cloern 1987), NH_4^+ may be preferred because the energy requirements for using this reduced N source are less than those for NO_3^- (Syrett 1981; Molloy and Syrett 1988; Dortch 1990). In light-limited waters, motile phytoplankton groups (e.g., dinoflagellates, cryptomonads) are capable of migrating to near surface depths, ensuring access to radiant energy needed to reduce NO_3^- to NH_4^+ , a critical step for incorporating this oxidized form of N into biosynthetic pathways and growth. Non-motile taxa must cope with deeper, lower irradiance waters, possibly limiting NO_3^- uptake and thus NH_4^+ uptake. Intrinsic physiological differences in N uptake among different phytoplankton taxa exist (e.g., Eppley et al. 1969; Stolte et al. 1994); these may lead to contrasting taxonomic responses to different N sources (Van Rijn et al. 1986; Collos 1989; Riegman 1998). Under conditions of restricted N availability, characteristic of many N-limited estuaries, such differences can lead to intense competition for either NH_4^+ , NO_3^- , or ON.

Bioassay experiments have shown that major estuarine phytoplankton functional groups (e.g., diatoms, dinoflagellates, cryptomonads, cyanobacteria, chlorophytes) may exhibit different growth responses to varying N sources and mixtures (Harrington 1999). Differential responses are not consistent in time and space. Other complex environmental factors, including light availability, water column mixing depth, water residence time, and temperature interact with phytoplankton community N uptake dynamics and growth rates (Cloern 1987, 1996, 1999; Richardson et al. 2001). Peierls and Paerl (1997) and Seitzinger and Sanders (1999) showed that atmospherically-derived DON also stimulated bacterial and phytoplankton growth. Atmospherically derived DON may selectively stimulate growth of facultative heterotrophic and photoheterotrophic phytoplankton taxa (e.g., dinoflagellates and cyanobacteria; Neilson and Lewin 1974; Antia et al. 1991; Paerl 1991).

Taxa-selective phytoplankton responses to specific N inputs may induce specific changes at the zooplankton, herbivorous fish, invertebrate, and higher consumer levels (i.e., bottom up effect). Shifts in phytoplankton community composition may also alter the flux of carbon, N, phosphorus (P), and other nutrients as well as oxygen in the estuary (top down effect). If poorly grazed, bloom-forming phytoplankton species dominate nutrient utilization and growth, their biomass is more likely to be deposited in the sediments than transferred up the food chain. This may enhance sediment oxygen consumption, hypoxia, and anoxia (Fig. 6). Readily consumed (grazed) phytoplankton species will more effectively support production at higher trophic levels, promoting both export (via migratory fish) and respiration of their biomass in the water column, thereby reducing sediment oxygen demand. These differential pathways and fates of primary production affect estuarine nutrient cycling, water quality, and habitability. Inedible, sedimented phytoplankton blooms will, by virtue of exacerbating hypoxia potentials, lead to enhanced

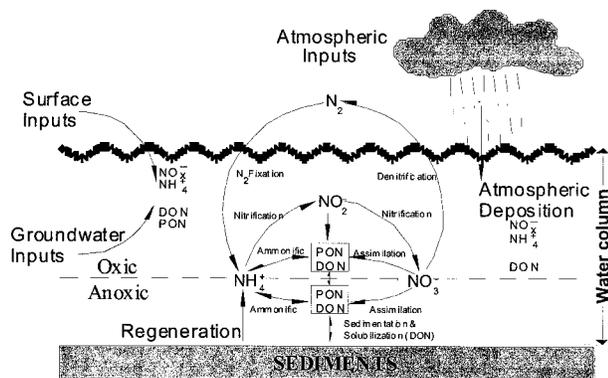


Fig. 5. Conceptual diagram illustrating the role of atmospheric deposition of nitrogen in estuarine nitrogen cycling.

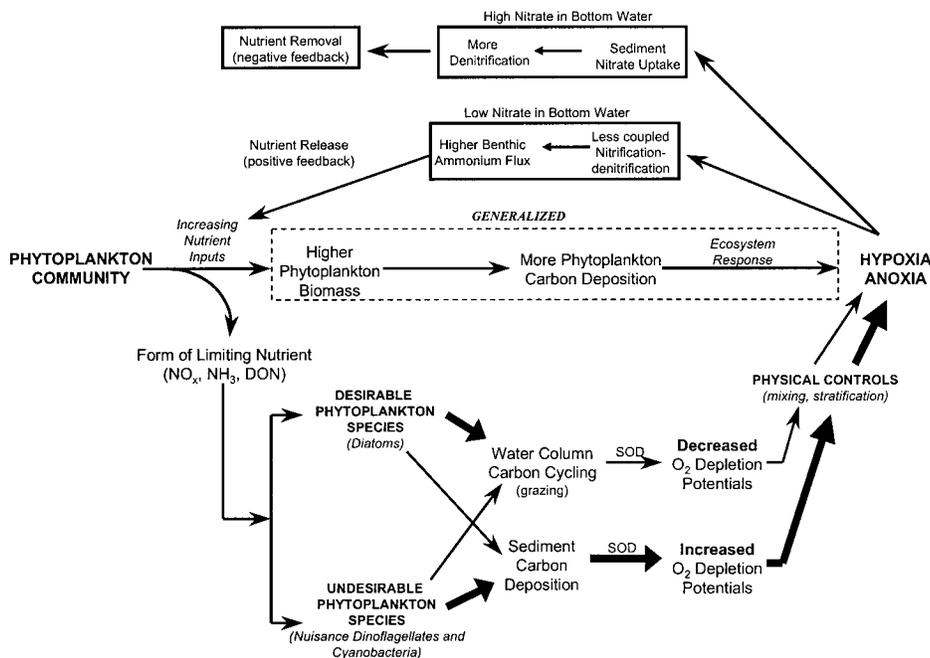


Fig. 6. Conceptual diagram illustrating the impacts of changes in phytoplankton composition on estuarine carbon flux, sediment oxygen demand, and nutrient cycling in response to eutrophication. If an increase in externally-supplied or new nutrient inputs lead to selective stimulation of phytoplankton that are not readily grazed and transferred up the food web (e.g., nuisance cyanobacteria and dinoflagellates), these phytoplankton will proportionately increase the sediment oxygen demand (SOD) and hypoxia and anoxia burden of the estuary. By contrast, if readily-consumed phytoplankton are favored, SOD will be reduced.

potentials may be affected. In poorly flushed estuaries this feedback loop could support bloom persistence by ensuring continuing nutrient regeneration. This scenario is promoted by warm, vertically stratified, long residence time conditions.

Due to changes in emission sources (i.e., agricultural, urban, industrial), certain forms of AD-N are increasing relative to others. One example is intensive animal (poultry, swine, cattle) operations in western Europe and the U.S. mid-Atlantic re-

gion, which have led to elevated ammonium deposition rates (Holland et al. 1999). In the U.S., depositional changes are reflected in long-term (since the mid to late 1970s) data from the network of NADP sites including one located in Sampson County, coastal North Carolina (NC-35), a region supporting a high density of industrial-style hog farms (Fig. 7). This location illustrates a precipitous rise in annual NH₄⁺ deposition. In addition, NH₄⁺ deposition has increased relative to NO₃⁻ deposition (Fig. 7), reflecting a national trend in agricultural regions (e.g., upper Midwest). The increasing ratio of NH₄⁺ to NO₃⁻ deposition reflects the rapid growth in intensive animal operations and simultaneous controls on NO_x emissions (NADP 2000).

Increases in the ratios of NH₄⁺ to NO₃⁻ as well as other AD-N sources may result in community compositional changes, because different forms of N may be differentially used by phytoplankton and other microorganisms. We are concerned about the amounts and composition of AD-N inputs because they can impact algal production and community composition, translating into differential water quality impacts (e.g., harmful versus non-harmful algal blooms).

It might be argued that the form in which new N enters an estuary is irrelevant because it may be

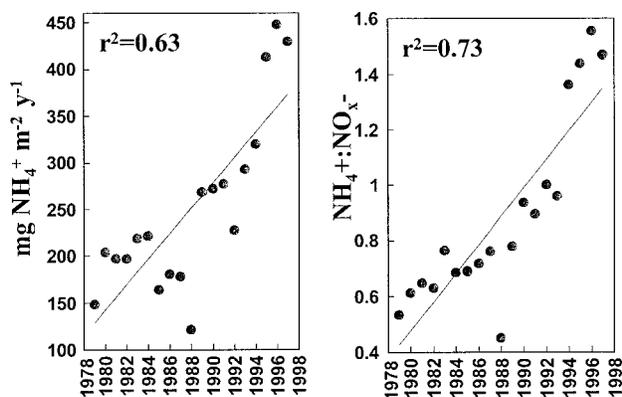


Fig. 7. A 20-year record (1978–1998) of NH₄⁺ and the ratio of NH₄⁺ to NO₃⁻ in wet deposition at NADP site NC 35, Sampson County, North Carolina (data from Paerl and Whitall 1999).

transformed via in-system N cycling before it is utilized by phytoplankton. In most N-limited estuarine and coastal systems, new N inputs are very rapidly (within hours in the Neuse River estuary) assimilated by resident N-starved phytoplankton (Pinckney et al. 1999). Large episodic inputs of new N, such as those delivered by rainstorms, may be rapidly used by resident phytoplankton (Pinckney et al. 1999). The chemical form in which this new N is delivered may determine phytoplankton community and ecosystem responses.

Harrington (1999) experimentally examined phytoplankton community response at the group level to different forms of N under different irradiance levels. Bioassays were designed in which water samples from the Neuse River estuary were amended with equimolar amounts of N in different forms (ammonium-only, nitrate-only, urea-only, and combined ammonium, nitrate, and urea) then either incubated at ambient irradiance or shaded to 10% of ambient irradiance. High performance liquid chromatography-based diagnostic photopigment analyses were used to characterize phytoplankton community responses, including the relative abundance of major algal groups (diatoms, dinoflagellates, cryptomonads, cyanobacteria, chlorophytes). Bioassays performed in the spring of 1998 showed that different forms of N caused community shifts at both 100% and 10% of ambient irradiance, reflecting the range of natural light conditions in the estuary. Results indicated that additions of different forms of N result in shifts in phytoplankton community composition; these shifts were also reliant on irradiance regimes (Fig. 8). The implications of such shifts for food web alterations and harmful algal bloom potentials are under further investigation.

Enrichment of N relative to other essential nutrients also alters the stoichiometric ratios and availabilities of those nutrients. In the Chesapeake Bay and its tributaries, excessive anthropogenic N loading delivered during spring high runoff freshet is a chief causative agent for periods of P limitation and co-limitation (Boynton et al. 1995). In the reduced runoff summer months, when N loading decreases and denitrification assumes an important N loss term, productivity in this system is more exclusively N limited. Studies in the Mississippi plume region of the northern Gulf of Mexico (Rabalais et al. 1996) demonstrate the impacts of excessive N enrichment from the Mississippi River on eutrophication potentials and availability of other growth-limiting nutrients. The availability of silicon (Si), which is required by diatoms, has been strongly affected. While N enrichment has closely followed human expansion in the Mississippi basin, Si concentrations have decreased. N-driven eu-

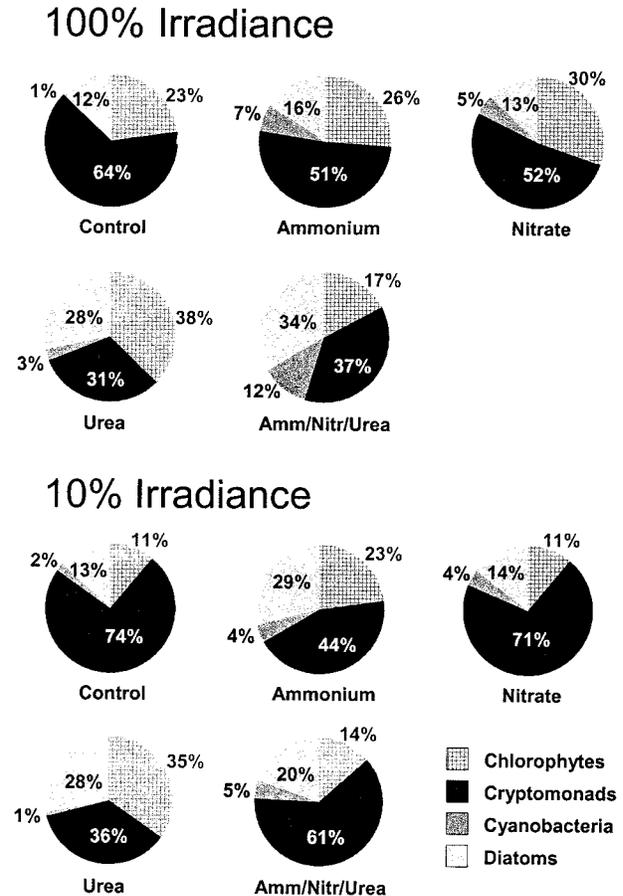


Fig. 8. Phytoplankton community composition in bioassays performed in April and May 1998. Nitrogen additions were equimolar, and identical sets of samples were incubated at 100% and 10% of ambient irradiance under natural irradiance and temperature conditions. The pie represents the total amount of chlorophyll *a*, which is partitioned among the major algal groups (adapted from Harrington 1999).

trophy has increased demands of Si, much of which is biogenically sedimented. Si resupply rates have not kept up with productivity demands, causing increased Si limitation and decline in diatom dominance. This impacts grazing and higher trophic levels (fisheries), which are dependent on diatom production. Atmospheric deposition, being enriched in N relative to Si (or P) would have a tendency to exacerbate such specific nutrient limitations and potential phytoplankton community shifts.

Atmospheric deposition may also be enriched with metals, either originating from dust, industrial effluents or volcanism (Church et al. 1984; Duce 1986). Trace metal enrichment (especially iron, Fe) in AD and other anthropogenic sources (dust, industrial emissions) may synergistically interact with N to stimulate coastal production and blooms

(Takeda et al. 1995; Zhuang et al. 1995; Paerl et al. 1999). This synergistic effect may exceed that observed with N alone, making atmospheric deposition a particularly potent stimulant of marine primary production (Paerl et al. 1999). That atmospheric-deposition of N, Fe, and other nutrients is a significant source of new nutrients extending seaward beyond estuarine nutrient stripping zones creates a basis for suspecting this source to play key roles in coastal eutrophication and harmful algal bloom dynamics (Zhang 1994; Paerl 1997).

Research and Management Implications

AD-N warrants close scrutiny and management from water quality, habitability, and fisheries resource perspectives. Management of N emissions, transport, and deposition must be incorporated in air/watershed and larger-scale nutrient management schemes. The scale of AD-N management to be considered in combination with watershed management is geographically quite encompassing, as suggested by the overlap of the oxidized-N airsheds of widely-spaced estuaries (Fig. 2) and the broad spatial coverage of the reduced-N airsheds (Fig. 3). In the case of oxidized airsheds for the mid-Atlantic region there is overlap with at least six contiguous watersheds (Fig. 9). A super airshed is defined as the outer envelope of the oxidized-N airshed of the first 11 watersheds in Table 1 (New England and mid-Atlantic region). The percent of oxidized-N deposition explained by this super airshed is greater than 80% for a majority of the 11 watersheds. This super region for the East Coast covers 20 states of the eastern U.S. and overlaps part of the eastern Mississippi drainage area. A similar enhancement of the percent oxidized-N deposition explained occurs for the Gulf Coast estuaries with the construction of a super airshed based on the Gulf Coast and Southeast airsheds. It would appear very difficult for a watershed or state to have a major effect on AD-N if acting alone. Efforts to address coastal AD-N management need to be as large as the multi-state regional (and national) air pollution programs for ozone and fine particulates spawned by the 1990 Clean Air Act Amendments to be efficient and effective.

ACKNOWLEDGMENTS

We appreciate the technical assistance of M. Go, B. Hendrickson, B. Peierls, L. Twomey, and P. Wyrick. J. Pinckney kindly provided illustrative materials. This work was supported by the National Science Foundation (DEB 9815495), U.S. Department of Agriculture NRI Project 9600509, U.S. Environmental Protection Agency STAR Project R82-5243-010, National Oceanic and Atmospheric Administration/North Carolina Sea Grant Program R/MER-43, The Environmental Defense Fund (Graduate Fellowship to D. R. Whitall), and the North Carolina Department of Natural Resources and Community Development, Division of Air Quality and Division of Water Quality/University

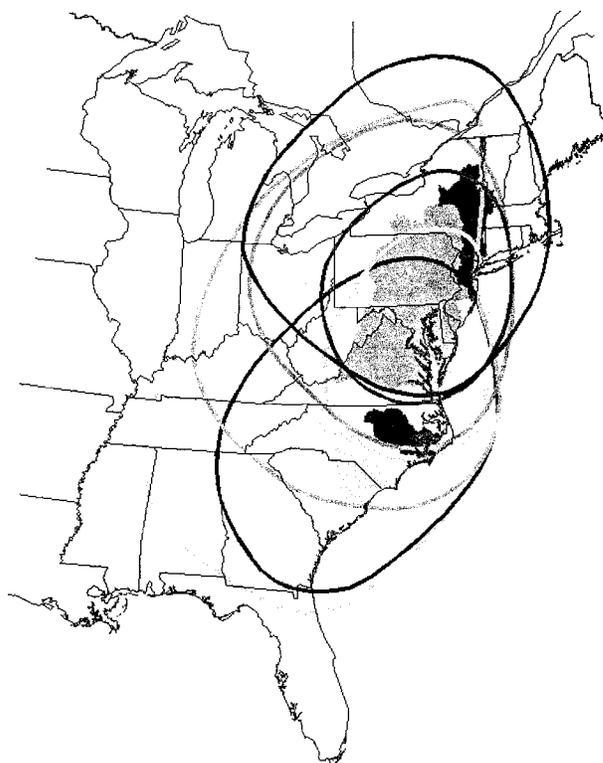


Fig. 9. Oxidized-nitrogen airsheds that significantly overlap and whose outer boundary comprises a northeast super-region: Hudson/Raritan Bay, Barnegat Bay, Delaware Bay, Delaware Inland Bays, Chesapeake Bay, Pamlico Sound, and Winyah Bay.

of North Carolina Water Resources Research Institute (Neuse River Estuary Monitoring and Modeling Project-ModMon). This paper has been reviewed in accordance with the U.S. Environmental Protection Agency's peer review policies and approved for publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

LITERATURE CITED

- AMBIO. 1990. Marine eutrophication. *Ambio* 19:101-176.
- ANEJA, V. P., J. P. CHUAHAN, AND J. T. WALKER. 2000. Characteristics of atmospheric ammonia emissions from swine waste storage and treatment lagoons. *Journal of Geophysical Research* 105:11535-11545.
- ANTIA, N., P. HARRISON, AND L. OLIVEIRA. 1991. The role of dissolved organic nitrogen in phytoplankton nutrition, cell biology and ecology. *Phycologia* 30:1-89.
- ASMAN, W. 1994. Emission and deposition of ammonia and ammonium. *Nova Acta Leopoldina* 70:263-297.
- BOYER, J., D. STANLEY, AND R. CHRISTIAN. 1994. Dynamics of NH_4^+ and NO_3^- uptake in the water column of the Neuse River estuary, North Carolina. *Estuaries* 17:361-371.
- BOYNTON, W. R., J. H. GARBER, R. SUMMERS, AND W. M. KEMP. 1995. Inputs, transformations and transport of nitrogen and phosphorus in Chesapeake Bay and selected tributaries. *Estuaries* 18:285-314.
- BRICKER, S. B., C. G. CLEMENT, D. E. PIRHALLA, S. P. ORLANDO, AND D. G. G. FARROW. 1999. National estuarine eutrophication assessment: Effects of nutrient enrichment in the nation's estuaries. Special Projects Office and the National Centers for Coastal Ocean Science. National Ocean Service, Na-

- tional Oceanic and Atmospheric Administration, Silver Spring, Maryland.
- BROOK, J. R., P. J. SAMSON, AND S. SILLMAN. 1995a. Aggregation of selected three-day periods to estimate annual and seasonal wet deposition totals for sulfate, nitrate, and acidity. Part I: A synoptic and chemical climatology for eastern North America. *Journal of Applied Meteorology* 34:297-325.
- BROOK, J. R., P. J. SAMSON, AND S. SILLMAN. 1995b. Aggregation of selected three-day periods to estimate annual and seasonal wet deposition totals for sulfate, nitrate, and acidity. Part II: Selection of events, deposition totals, and source-receptor relationships. *Journal of Applied Meteorology* 34:326-339.
- CASTRO, M. S., C. T. DRISCOLL, T. E. JORDAN, W. R. REAY, W. R. BOYNTON, S. P. SEITZINGER, R. V. STYLES, AND J. E. CABLE. 2000. Contribution of atmospheric deposition to the total nitrogen loads of thirty-four estuaries on the Atlantic and Gulf Coast of the United States, p. 77-106. In R. Valigura (ed.), *Atmospheric Nitrogen Deposition in Coastal Waters*. Coastal Estuarine Science Series No. 57. American Geophysical Union Press, Washington, D.C.
- CHANG, J., R. BROST, I. ISAKSEN, S. MADRONICH, P. MIDDLETON, W. STOCKWELL, AND C. WALCEK. 1987. A three-dimensional Eulerian acid deposition model: Physical concepts and formulation. *Journal of Geophysical Research* 92:14681-14700.
- CHESAPEAKE BAY PROGRAM. 1994. Annual report #2 on atmospheric N deposition: 1990-1994. Chesapeake Bay Program, U.S. Environmental Protection Agency, Annapolis, Maryland.
- CHRISTIAN, R. R., J. N. BOYER, AND D. W. STANLEY. 1991. Multi-year distribution patterns of nutrients within the Neuse River estuary. *Marine Ecology Progress Series* 71:259-274.
- CHURCH, T. M., J. M. TRAMONTANTO, J. R. SCUDLARK, T. D. JICKELLS, J. J. TOKOS, AND A. H. KNAPP. 1984. The wet deposition of trace metals to the western Atlantic Ocean at the mid-Atlantic coast and Bermuda. *Atmospheric Environment* 18:2657-2664.
- CLARKE, J., E. EDGERTON, AND B. MARTIN. 1997. Dry deposition calculations for the Clean Air Status and Trends Network. *Atmospheric Environment* 31:3667-3678.
- CLOERN, J. E. 1987. Turbidity as a control on phytoplankton biomass and productivity in estuaries. *Continental Shelf Research* 7:1367-1382.
- CLOERN, J. E. 1996. Phytoplankton bloom dynamics in coastal ecosystems: A review with general lessons from sustained investigations of San Francisco Bay, California. *Reviews in Geophysics* 34:127-168.
- CLOERN, J. E. 1999. The relative importance of light and nutrient limitation of phytoplankton growth: A simple index of coastal ecosystems sensitivity to nutrient enrichment. *Aquatic Ecology* 33:3-16.
- COHN, R. AND R. DENNIS. 1994. The evaluation of acid deposition models using principal component spaces. *Atmospheric Environment* 28:2531-2543.
- COLLOS, Y. 1989. A linear model of external interactions during uptake of different forms of inorganic nitrogen by microalgae. *Journal of Plankton Research* 11:521-533.
- COPELAND, B. J. AND J. GRAY. 1991. Status and trends report of the Albemarle-Pamlico estuary. Albemarle-Pamlico Estuarine Study Report 90-01. North Carolina Department of Environmental Health and Natural Resources, Raleigh, North Carolina.
- CORNELL, S., A. RENDELL, AND T. D. JICKELLS. 1995. Atmospheric inputs of dissolved organic nitrogen to the oceans. *Nature* 376:243-246.
- CORRELL, D. AND D. FORD. 1982. Comparison of precipitation and land runoff as sources of estuarine nitrogen. *Estuarine and Coastal Shelf Science* 15:45-56.
- DAVIDSON, C. L. AND L. WU. 1990. Dry deposition of particle and vapors, p. 103-209. In S. E. Lindberg, A. L. Page, and S. A. Norton (eds.), *Acidic Precipitation, Sources, Deposition and Canopy Interaction*. Springer, New York.
- D'ELIA, C. F., J. G. SANDERS, AND W. R. BOYNTON. 1986. Nutrient enrichment studies in a coastal plain estuary: Phytoplankton growth in large scale, continuous cultures. *Canadian Journal of Fisheries and Aquatic Sciences* 43:397-406.
- DENNIS, R. 1997. Using the regional acid deposition model to determine the nitrogen deposition airshed of the Chesapeake Bay watershed, p. 393-413. In J. E. Baker (ed.), *Atmospheric Deposition of Contaminants to the Great Lakes and Coastal Waters*. Society of Environmental Toxicology and Chemistry Press, Pensacola, Florida.
- DENNIS, R. L., F. S. BINKOWSKI, T. L. CLARK, J. N. MCHENRY, S. J. REYNOLDS, AND S. K. SEILKOP. 1990. Selected applications of the regional acid deposition model and engineering model, Appendix 5F (Part 2) of NAPAP SOS/T Report 5. In P. M. Irving (ed.), *National Acid Precipitation Assessment Program: State of Science and Technology, Volume 1. National Acid Precipitation Assessment Program*, Washington, D.C.
- DIAZ, R. AND R. ROSENBERG. 1995. Marine benthic hypoxia: A review of its ecological effects and the behavioral responses of benthic macrofauna. *Oceanography and Marine Biology Annual Review* 33:245-303.
- DORTCH, Q. 1990. The interaction between ammonium and nitrate uptake in phytoplankton. *Marine Ecology Progress Series* 61:183-201.
- DUCE, R. 1986. The impact of atmospheric nitrogen, phosphorus, and iron species on marine biological productivity, p. 497-529. In P. Buat-Menard (ed.), *The Role of Air-Sea Exchange in Geochemical Cycling*. D. Reidel, Norwell, Massachusetts.
- DUCE, R. 1991. Chemical exchange at the air-coastal sea interface, p. 91-110. In R. Mantoura, J. Martin, and R. Wollast (eds.), *Ocean Margin Processes in Global Change*. J. Wiley and Sons, Chichester, U.K.
- DUGDALE, R. C. 1967. Nitrogen limitation in the seas: Dynamics, identification, and significance. *Limnology and Oceanography* 12:685-695.
- DUYZER, J. 1994. Dry deposition of ammonia and ammonium aerosols over heathland. *Journal of Geophysical Research* 99:18757-18763.
- ECOLOGICAL SOCIETY OF AMERICA. 1998. *The Role of Atmospheric Deposition in Coastal Eutrophication*. Ecological Society of America Publications, Washington, D.C.
- ELMGREN, R. 1989. Man's impact on the ecosystem of the Baltic Sea; Energy flows today and at the turn of the century. *Environmental Science and Technology* 9:635-638.
- ENVIRONMENTAL PROTECTION AGENCY. 1989. *Saving Bays and Estuaries: A Primer for Establishing and Managing Estuary Programs*. Office of Marine and Estuarine Protection, Washington, D.C.
- ENVIRONMENTAL PROTECTION AGENCY. 1996. *Air quality criteria for ozone and related photochemical oxidants*. Report EPA/600/P-93/004aF, Volume I of III. Office of Research and Development, Washington, D.C.
- ENVIRONMENTAL PROTECTION AGENCY. 1999. *Deposition of air pollutants to the great waters*. Third Report to Congress. U.S. Government Printing Office, Washington, D.C.
- EPPLEY, R. W., J. N. ROGERS, AND J. J. MCCARTHY. 1969. Half saturation constants for uptake of nitrate and ammonia by marine phytoplankton. *Limnology and Oceanography* 14:912-920.
- FISHER, D. AND M. OPPENHEIMER. 1991. Atmospheric nitrogen deposition and the Chesapeake Bay estuary. *Ambio* 20:102-108.
- FOWLER, D., C. FLECHARD, M. SUTTON, AND R. STORETON-WEST. 1998. Long term measurements of the land-atmosphere exchange of ammonia over moorland. *Atmospheric Environment* 32:453-459.

- GALLOWAY, J., H. LEVY, AND P. KASIBHATIA. 1994. Year 2020: Consequences of population growth and development on deposition of oxidized nitrogen. *Ambio* 23:120–123.
- GOOLSBY, D. A., W. A. BATTAGLIN, G. B. LOWRANCE, R. S. ARTZ, B. J. AULENBACH, AND R. P. HOOPER. 1999. Flux and sources of nutrients in the Mississippi-Atchafalaya River basin. Topic 3 for the Integrated Assessment of Hypoxia in the Gulf of Mexico. National Oceanic and Atmospheric Administration Coastal Ocean Program Decision Analysis No. 17. National Oceanic and Atmospheric Administration Coastal Ocean Program, Silver Spring, Maryland.
- GROUP OF EXPERTS ON THE SCIENTIFIC ASPECTS OF MARINE POLLUTION (GESAMP). 1989. The atmospheric input of trace species to the world ocean: Report and Studies No. 38. World Meteorological Association, Geneva.
- HARRINGTON, M. B. 1999. Responses of natural phytoplankton communities from the Neuse River estuary, North Carolina to changes in nitrogen supply and incident irradiance. M.Sc. Thesis, University of North Carolina at Chapel Hill, Chapel Hill, North Carolina.
- HINGA, K. R., A. A. KELLER, AND C. A. OVIATT. 1991. Atmospheric deposition and nitrogen inputs to coastal waters. *Ambio* 20: 256–260.
- HOBBIE, J. E. AND N. W. SMITH. 1975. Nutrients in the Neuse River estuary, North Carolina, Report No. UNC-SG-75-21. UNC Sea Grant Program, North Carolina State University, Raleigh, North Carolina.
- HOLLAND, E., F. DENTENER, B. BRASWELL, AND J. SULZMAN. 1999. Contemporary and pre-industrial global reactive nitrogen budgets. *Biogeochemistry* 43:7–43.
- HOV, Ø. AND B. A. HJØLLO. 1994. Transport distance of ammonia and ammonium in northern Europe I. Its relation to emissions of SO₂ and NO_x. *Journal of Geophysical Research* 99: 18749–18755.
- HOV, Ø., B. A. HJØLLO, AND A. ELIASSEN. 1994. Transport distance of ammonia and ammonium in northern Europe I. Model description. *Journal of Geophysical Research* 99:18735–18748.
- HOWARTH, R. W. 1998. An assessment of human influences on inputs of nitrogen to the estuaries and continental shelves of the North Atlantic Oceans. *Nutrient Cycling in Agroecosystems* 52: 213–223.
- HOWARTH, R. W., G. BILLEN, D. SWANEY, A. TOWNSEND, N. JAWORSKI, K. LAJTHA, J. A. DOWNING, R. ELMGREN, N. CARACO, T. JORDAN, F. BERENDSE, J. FRENEY, V. KUDEYAROV, P. MURDOCH, AND Z. ZHAO-LIANG. 1996. Regional nitrogen budgets and riverine N and P fluxes for the drainages to the North Atlantic Ocean: Natural and human influences. *Biogeochemistry* 35:75–139.
- JAWORSKI, N., R. HOWARTH, AND L. HETLING. 1997. Atmospheric deposition of nitrogen oxides onto the landscape contributes to coastal eutrophication in the northeast United States. *Environmental Science and Technology* 31:1995–2004.
- JUSTIC, D., N. N. RABALAIS, R. E. TURNER, AND W. J. WISEMAN, JR. 1993. Seasonal coupling between riverborne nutrients, net productivity and hypoxia. *Marine Pollution Bulletin* 26:184–189.
- KEENE, W. C. AND D. L. SAVOIE. 1998. The pH of deliquesced sea-salt aerosol in polluted marine air. *Geophysical Research Letters* 25:2181–2184.
- KENNEDY, V. 1983. The Estuary as a Filter. Academic Press, New York.
- KUENZLER, E. J., D. W. STANLEY, AND J. P. KOENINGS. 1979. Nutrient kinetics of phytoplankton in the Pamlico River, North Carolina. University of North Carolina Water Resources Research Institute Report No. 139. UNC Water Resources Institute, Raleigh, North Carolina.
- LANGFORD, A. AND F. FEHSENFELD. 1992. Natural vegetation as a source or sink for atmospheric ammonia: A case study. *Science* 255:581–583.
- LIKENS, G., F. BORMAN, AND M. JOHNSON. 1974. Acid rain. *Environment* 14:33–40.
- LONG ISLAND SOUND STUDY. 1996. Report on nitrogen and organic carbon loads to Long Island Sound 1996. Prepared by P. Stacey, Connecticut Department of Environmental Protection. Bureau of Water Management, Hartford, Connecticut.
- LUKE, W. AND R. DICKERSON. 1987. Flux of reactive nitrogen compounds from eastern North America to the western Atlantic Ocean. *Global Biogeochemical Cycles* 1:329–343.
- MARTIN, J. M., F. ELBAZ-POULICHET, C. GWUE, M. D. LOYE-PILOT, AND G. HAN. 1989. River versus atmospheric input of material to the Mediterranean Sea: An overview. *Marine Chemistry* 28: 159–182.
- MATHUR, R. AND R. L. DENNIS. 2000. A regional modeling analysis of reduced nitrogen cycling in the eastern United States, p. 85–88. In Conference Proceedings: American Meteorological Society, Preprints of the Symposium on Atmospheric Chemistry: Issues in the 21st Century, 9–14 January 2000, Long Beach California. American Meteorological Society, Boston, Massachusetts.
- MEYERS, T., P. FINKELSTEIN, J. CLARKE, T. ELLESTAD, AND P. SIMS. 1998. A multilayer model for inferring dry deposition using standard meteorological measurements. *Journal of Geophysical Research* 103:22645–22661.
- MOLLOY, C. AND P. SYRETT. 1988. Interrelationships between uptake of urea and uptake of ammonium by microalgae. *Journal of Experimental Marine Biology and Ecology* 118:85–95.
- MOPPER, K. AND R. ZIKA. 1987. Free amino acids in marine rains: Evidence for oxidation and potential role in nitrogen cycling. *Nature* 325:246–249.
- MOSER, F. C. 1997. Sources and sinks of nitrogen and trace metals, and benthic macrofauna assemblages in Barnegat Bay, New Jersey. Ph.D. Dissertation, Rutgers, The State University of New Jersey, New Brunswick, New Jersey.
- NATIONAL ACID DEPOSITION PROGRAM. 2000. 1999 Annual Summary of the NADP Program. Illinois State Water Survey, Champaign, Illinois.
- NATIONAL RESEARCH COUNCIL. 2000. Clean Coastal Waters: Understanding and Reducing the Effects of Nutrient Pollution. National Academy Press, Washington, D.C.
- NEILSON, A. H. AND R. A. LEWIN. 1974. The uptake and utilization of organic carbon by algae: An essay in comparative biochemistry. *Phycologia* 13:227–264.
- NEILSON, B. AND L. CRONIN (EDS.). 1981. Estuaries and Nutrients. Humana Press, Clifton, New Jersey.
- NIXON, S. W. 1981. Remineralization and nutrient cycling in coastal marine ecosystems, p. 111–138. In B. J. Neilson and L. E. Cronin (eds.), Estuaries and Nutrients. Humana Press, Clifton, New Jersey.
- NIXON, S. W. 1986. Nutrient dynamics and the productivity of marine coastal waters, p. 91–115. In D. Clayton and M. Behbehani (eds.), Coastal Eutrophication. The Alden Press, Oxford, U.K.
- NIXON, S. W. 1995. Coastal marine eutrophication: A definition, social causes, and future concerns. *Ophelia* 41:199–220.
- NIXON, S. W., J. W. AMMERMAN, L. P. ATKINSON, V. M. BEROUNSKY, G. BILLEN, W. C. BOICOURT, W. R. BOYNTON, T. M. CHURCH, D. M. DITORO, R. ELMGREN, J. H. GARBER, A. E. GIBLIN, R. A. JAHNKE, N. J. P. OWENS, M. E. Q. PILSON, AND S. P. SEITZINGER. 1996. The fate of nitrogen and phosphorus at the land-sea margin of the North Atlantic Ocean. *Biogeochemistry* 35:141–180.
- PAERL, H. W. 1983. Factors regulating nuisance blue-green algal bloom potentials in the lower Neuse River. Report No. 177. University of North Carolina Water Resources Research Institute, Raleigh, North Carolina.
- PAERL, H. W. 1985. Enhancement of marine primary production by nitrogen-enriched acid rain. *Nature* 316:747–749.
- PAERL, H. W. 1988. Nuisance phytoplankton blooms in coastal,

- estuarine, and inland waters. *Limnology and Oceanography* 33: 823–847.
- PAERL, H. W. 1991. Ecophysiological and trophic implications of light-stimulated amino acid utilization in marine picoplankton. *Applied and Environmental Microbiology* 57:473–479.
- PAERL, H. W. 1995. Coastal eutrophication in relation to atmospheric nitrogen deposition: Current perspectives. *Ophelia* 41: 237–259.
- PAERL, H. W. 1997. Coastal eutrophication and harmful algal blooms: Importance of atmospheric deposition and groundwater as “new” nitrogen and other nutrient sources. *Limnology and Oceanography* 42:1154–1165.
- PAERL, H. W. AND M. L. FOGEL. 1994. Isotopic characterization of atmospheric nitrogen inputs as sources of enhanced primary production in coastal Atlantic Ocean waters. *Marine Biology* 119:635–645.
- PAERL, H. W., M. A. MALLIN, C. DONAHUE, M. GO, AND B. L. PEIERLS. 1995. Nitrogen loading sources and eutrophication of the Neuse River estuary, North Carolina: Direct and indirect roles of atmospheric deposition. Report 291, University of North Carolina Water Resources Research Institute, Raleigh, North Carolina.
- PAERL, H., J. PINCKNEY, J. FEAR, AND B. L. PEIERLS. 1998. Ecosystem responses to internal and watershed organic matter loading: Consequences for hypoxia in the eutrophying Neuse River estuary, North Carolina. *Marine Ecology Progress Series* 166: 17–25.
- PAERL, H. W. AND D. R. WHITALL. 1999. Anthropogenically-derived atmospheric nitrogen deposition, marine eutrophication and harmful algal bloom expansion: Is there a link? *Ambio* 28:307–311.
- PAERL, H. W., J. D. WILLEY, M. GO, B. L. PEIERLS, J. L. PINCKNEY, AND M. L. FOGEL. 1999. Rainfall stimulation of primary production in western Atlantic Ocean waters: Roles of different nitrogen sources and co-limiting nutrients. *Marine Ecology Progress Series* 176:205–214.
- PEIERLS, B. L., N. F. CARACO, M. L. PACE, AND J. J. COLE. 1991. Human influence on river nitrogen. *Nature* 350:386–387.
- PEIERLS, B. L. AND H. W. PAERL. 1997. The bioavailability of atmospheric organic nitrogen deposition to coastal phytoplankton. *Limnology and Oceanography* 42:1819–1880.
- PINCKNEY, J. L., H. W. PAERL, AND M. B. HARRINGTON. 1999. Responses of the phytoplankton community growth rate to nutrient pulses in variable estuarine environments. *Journal of Phycology* 35:1455–1463.
- PRADO-FIEDLER, R. R. 1990. Atmospheric input of inorganic nitrogen species to the Kiel Bight. *Helgolander Meeresuntersuchungen* 44:21–30.
- PROSPERO, J. M., K. BARRETT, T. CHURCH, F. DENTENER, R. A. DUCE, J. N. GALLOWAY, H. LEVY II, J. MOODY, AND P. QUINN. 1996. Atmospheric deposition of nutrients to the North Atlantic Basin. *Biogeochemistry* 35:27–73.
- PRYOR, S. C., R. J. BARTHELMIE, L. L. S. GEERNAERT, T. ELLERMANN, AND K. D. PERRY. 1999. Speciated particle dry deposition to the sea surface: Results from ASEPS 97. *Atmospheric Environment* 33:2045–2058.
- RABALAIS, N. N., R. E. TURNER, D. JUSTIC, Q. DORTCH, AND W. J. WISEMAN, JR. 1999. Characterization of Hypoxia. Topic 1 Report for the Integrated Assessment of Hypoxia in the Gulf of Mexico. National Oceanic and Atmospheric Administration Coastal Ocean Program Decision Analysis Series No. 15. NOAA Coastal Ocean Program, Silver Spring, Maryland.
- RABALAIS, N. N., R. E. TURNER, D. JUSTIC, Q. DORTCH, W. J. WISEMAN, JR., AND B. K. SEN GUPTA. 1996. Nutrient changes in the Mississippi River and system responses on the adjacent continental shelf. *Estuaries* 19:386–407.
- RICHARDSON, K. 1997. Harmful or exceptional phytoplankton blooms in the marine ecosystem. *Advances in Marine Biology* 31:302–385.
- RICHARDSON, T. L., J. L. PINCKNEY, AND H. W. PAERL. 2001. Responses of estuarine phytoplankton communities to nitrogen form and mixing using microcosm bioassays. *Estuaries* 24:828–839.
- RIEGMAN, R. 1998. Species composition of harmful algal blooms in relation to macronutrient dynamics, p. 475–488. In D. M. Anderson, A. D. Cembella, and G. M. Hallegraeff (eds.), *Physiological Ecology of Harmful Algal Blooms*. North Atlantic Treaty Organization Series Volume G 41, Springer, Berlin, Germany.
- RIZZO, W., G. LACKEY, AND R. CHRISTIAN. 1992. Significance of euphotic, subtidal sediments to oxygen and nutrient cycling in a temperate estuary. *Marine Ecology Progress Series* 86:51–61.
- RUDEK, J., H. W. PAERL, M. A. MALLIN, AND P. W. BATES. 1991. Seasonal and hydrological control of phytoplankton nutrient limitation in the lower Neuse River estuary, North Carolina. *Marine Ecology Progress Series* 75:133–142.
- RUSSELL, K., J. GALLOWAY, S. MACKO, J. MOODY, AND J. SKUDLARK. 1998. Sources of nitrogen in wet deposition to the Chesapeake Bay region. *Atmospheric Environment* 32:2453–2465.
- RYTHER, J. AND W. DUNSTAN. 1971. Nitrogen, phosphorus and eutrophication in the coastal marine environment. *Science* 171:1008–1112.
- SEINFELD, J. AND S. PANDIS. 1998. *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*. John Wiley & Sons, Inc., New York.
- SEITZINGER, S. P. 1988. Denitrification in freshwater and coastal marine ecosystems: Ecological and geochemical significance. *Limnology and Oceanography* 33:702–724.
- SEITZINGER, S. P. AND R. W. SANDERS. 1999. Atmospheric inputs of organic nitrogen stimulate estuarine bacteria and phytoplankton. *Limnology and Oceanography* 44:721–730.
- SKUDLARK, J. AND T. CHURCH. 1993. Atmospheric input of inorganic nitrogen to Delaware Bay. *Estuaries* 16:747–759.
- SLINN, W. G. N. 1982. Predictions for particle deposition to vegetative canopies. *Atmospheric Environment* 16:1785–1794.
- SORTEBERG, A., Ø. HOV, S. SOLBERG, K. TØRSETH, H. ARESKOU, M. FERM, K. GRANBY, H. LÄTTILÄ, K. PERSSON, AND D. SIMPSON. 1998. Gaseous and particulate oxidized and reduced nitrogen species in the atmospheric boundary layer in Scandinavia in spring. *Journal of Atmospheric Chemistry* 30:241–271.
- STODDARD, J. L. 1993. Long-term changes in watershed retention of nitrogen: Its causes and aquatic consequences, p. 223–241. In L. A. Baker (ed.), *Environmental Chemistry of Lakes and Reservoirs*, Advances in Chemistry Series No. 237. American Chemical Society, Washington, D.C.
- STOLTE, W., T. MCCOLLIN, A. NOORDELOOS, AND R. RIEGMAN. 1994. Effects of nitrogen source on the size distribution within marine phytoplankton populations. *Journal of Experimental Marine Biology and Ecology* 184:83–97.
- SYRETT, P. J. 1981. Nitrogen metabolism of microalgae. *Canadian Bulletin of Fisheries and Aquatic Sciences* 210:182–210.
- TAKEDA, S., A. KAMATANI, AND K. KAWANOBE. 1995. Effects of nitrogen and iron enrichments on phytoplankton communities in the northwestern Indian Ocean. *Marine Chemistry* 50: 229–241.
- TAMPA BAY NATIONAL ESTUARY PROGRAM. 1996. *Charting the Course for Tampa Bay: Final Comprehensive Conservation and Management Plan*. Tampa Bay National Estuary Program, St. Petersburg, Florida.
- TIMPERLEY, M. R., R. VIGOR-BROWN, M. KAWASHIMA, AND M. ISHIGAMI. 1985. Organic nitrogen compounds in atmospheric precipitation: Their chemistry and availability to phytoplankton. *Canadian Journal of Fisheries and Aquatic Sciences* 42:1171–1177.
- TØRSETH, K. AND A. SEMB. 1998. Deposition of nitrogen and other major inorganic compounds in Norway, 1992–1996. *Environmental Pollution* 102:299–304.
- TURPIN, D. H. 1991. Effects of inorganic N availability on algal

- photosynthesis and carbon metabolism. *Journal of Phycology* 27:14–20.
- TYLER, M. 1988. Contributions of atmospheric nitrate deposition to nitrate loading in the Chesapeake Bay. VERSAR Inc. Maryland Department of Natural Resources Report RP1052. Annapolis, Maryland.
- VALIELA, I., K. FOREMAN, M. LAMONTAGNE, D. HERSH, J. COSTA, P. PECKOL, B. DEMEO-ANDERSON, C. D'AVANZO, M. BABIONE, C.-H. SHAM, J. BRAWLEY, AND K. LAJTHA. 1992. Couplings of watersheds and coastal waters: Sources and consequences of nutrient enrichment in Waquoit Bay, Massachusetts. *Estuaries* 15:443–457.
- VALIGURA, R. A., R. B. ALEXANDER, M. S. CASTRO, T. P. MEYERS, H. W. PAERL, P. E. STACEY, AND R. E. TURNER (EDS.). 2000. Nitrogen Loading in Coastal Water Bodies: An Atmospheric Perspective. Coastal and Estuarine Studies No. 57. American Geophysical Union, Washington, D.C.
- VALIGURA, R., W. LUKE, R. ARTZ, AND B. HICKS. 1996. Atmospheric Nutrient Inputs to Coastal Areas: Reducing the Uncertainties. U.S. National Oceanic and Atmospheric Administration Coastal Ocean Program Decision Analysis Series No. 9. Washington, D.C.
- VAN RIJN, J., M. SHILO, AND S. DIAB. 1986. Phytoplankton succession in relation to nitrogen regime in shallow, brackish-water fish ponds. *Archives of Hydrobiology* 111:183–195.
- VITOUSEK, P. M., H. A. MOONEY, J. LUBCHENKO, AND J. M. MELLILO. 1997. Human domination of Earth's ecosystems. *Science* 277:494–499.
- WHITALL, D. R. 2000. Atmospheric nitrogen deposition to the Neuse River watershed: Fluxes, sources and spatiotemporal variability. Ph.D. Dissertation, University of North Carolina at Chapel Hill, North Carolina.
- WHITALL, D. R. AND H. W. PAERL. 2001. Importance of atmospheric nitrogen deposition to the Neuse River estuary, North Carolina. *Journal of Environmental Quality* 30:1508–1515.
- WILLEY, J. D. AND H. W. PAERL. 1993. Enhancement of chlorophyll *a* production in Gulf Stream surface seawater by synthetic vs. natural rain. *Marine Biology* 116:329–334.
- WINN, R. AND D. KNOTT. 1992. An evaluation of the survival of experimental populations exposed to hypoxia in the Savannah River estuary. *Marine Ecology Progress Series* 88:161–179.
- WYERS, G. AND J. ERISMAN. 1998. Ammonia exchange over coniferous forest. *Atmospheric Environment* 32:441–451.
- ZHANG, J. 1994. Atmospheric wet deposition of nutrient elements: Correlation with harmful biological blooms in north-west Pacific coastal zones. *Ambio* 23:464–468.
- ZHUANG, G., Z. YI, AND G. T. WALLACE. 1995. Iron (II) in rainwater, snow, and surface seawater from a coastal environment. *Marine Chemistry* 50:41–50.

SOURCE OF UNPUBLISHED MATERIALS

MATHUR, R. Unpublished Data. Microelectronics Center of North Carolina, Environmental Modeling Center, 3021 Cornwallis Road, Research Triangle Park, North Carolina 27709.

Received for consideration, January 19, 2001

Accepted for publication, June 26, 2001