

itrogen Deposition

to Coastal

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Connecting Atmospheric

Deposition

to Coastal

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Research is needed to understand this air-water quality interaction.

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itrogen (N) is the nutrient recognized as the "currency" of estuarine and coastal plant production and water quality status. Although N inputs are essential for maintaining the fertility of N-sensitive waters, excessive loading has created "too much of a good thing". It can lead to habitat degradation, algal blooms, toxicity, hypoxia, anoxia, fish kills, and ultimately loss of biodiversity—all classic signs of eutrophication and the accompanying water quality and habitat degradation (1–3).

Urban, agricultural, and industrial expansion into coastal zones has been accompanied by a precipitous rise in N loading, which is most readily observed as Nenriched surface and subsurface discharge. When considering all of the anthropogenic N inputs to coastal waters, atmospheric deposition has previously been a neglected source. However, this perception is changing. Recent watershed- and regional-scale studies point to atmospheric deposition of nitrogen (AD-N) as a highly significant and growing source of externally supplied, or "new", N entering the coastal zone (4). During the past century, AD-N, much of which originates from combustion and agricultural emissions, has increased 10-fold and now accounts for more than 40% of new N-loading to coastal ecosystems (4). AD-N is both a local and a regional issue, because N emission sources may reside either within or far outside specific watersheds (5).

In this article, the scope of the AD-N problem is presented, some of the data are described, and an agenda for future work is outlined.

Consider the sources

AD-N provides the aquatic milieu with various biologically available N compounds that reflect diverse human

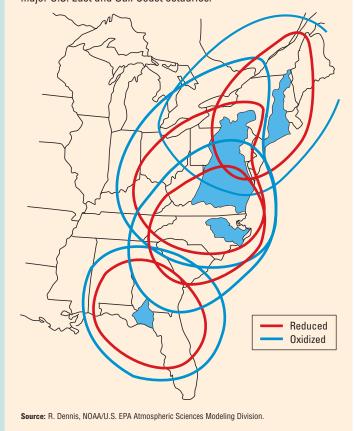
activities, including fossil fuel and biomass combustion, agricultural and industrial emissions, and to a lesser extent natural processes, such as volcanism and soil and water microbial volatilization. These compounds include inorganic reduced (ammonia/ ammonium), oxidized (nitrite/nitrate), and organic forms. Because AD-N ranges from 400 to more than 1200 mg N/m²yr, it is one of the largest sources of anthropogenic N delivered to North American and European coastal waters. On a larger scale, AD-N flux to the North Atlantic Ocean basin is at least 11 teragrams per year (Tg/yr) and accounts for 46-57% of the basin's new N, which surpasses North American and European riverine inputs of ~10 Tg/yr (6). Globally, AD-N contributes ~40 Tg/yr to the world's oceans, whereas total riverine inputs are on the order of 30 Tg N/yr and groundwater sources contribute another 10 Tg N/yr (6).

AD-N is important from both local watershed and larger-scale regional perspectives because the corresponding airshed extends far beyond the watershed boundary. The airshed is defined as the spatial range of N emission sources that contribute AD-N to specific water bodies. Figure 1 shows how a recent analy-

FIGURE 1

Principal nitrogen airsheds for Long Island Sound, Chesapeake Bay, Pamlico Sound, and Apalachee Bay

These comparative dimensions illustrate reduced and oxidized nitrogen sources for the watersheds and overlapping airsheds for major U.S. East and Gulf Coast estuaries.



sis of major estuarine ecosystems along the U.S. East and Gulf Coasts indicates that the area of the airsheds delivering reduced and oxidized N to these estuaries ranges from 10 to over 30 times greater than the area of the watersheds (5, 7). This means that AD-N originating from major metropolises such as Atlanta, Washington, D.C., Pittsburgh, Philadelphia, and Baltimore can impact remote N-sensitive estuarine systems, which have mostly rural agricultural watersheds, such as Albemarle-Pamlico Sound in North Carolina and Altamaha Sound in Georgia. Conversely, largely urban watersheds, including those surrounding Chesapeake and Delaware Bays, may be impacted by agriculturally derived N deposition from regions in southeastern states, such as North Carolina, South Carolina, and Virginia, which are far outside the affected watersheds.

Another unique and important aspect of AD-N is that rainfall and dry N deposition can directly affect estuarine and coastal waters downstream of regions where most of the terrestrially supplied N (wastewater, surface runoff, and groundwater) is effectively assimilated or filtered by N-limited microscopic suspended algae (phytoplankton), bacteria, and higher plants. Therefore, AD-N can "fertilize" waters downstream of the estuarine N "filtering zones". This may make AD-N a potentially important source of new N that could contribute to the formation of more distant coastal and oceanic algal blooms, including harmful red tides (8). In coastal regions bordering on the Baltic, North, western Mediterranean, and Yellow Seas, and the North American Atlantic Ocean, the ecological effects of AD-N on sensitive waters have been linked to various symptoms of eutrophication, including expanding algal blooms and changes in algal community compositions with corresponding food web alterations (2, 4, 9).

We must improve our knowledge of sources, routes, and fates of AD-N and its interaction with other essential nutrients and emission products, such as heavy metals, toxics, and organic compounds, to understand the impacts on coastal productivity, water quality, and food web dynamics. AD-N sources of interest include biologically available forms of dissolved inorganic nitrogen (DIN: nitrites and nitrates emitted from fossil fuel combustion and biomass burning; ammonia and ammonium volatilized from agricultural waste, fertilizers, decomposition, and biomass burning) and dissolved organic nitrogen (DON: sources unclear but thought to have both anthropogenic and natural origins). Increasing amounts of these forms of N reflect changing land use and human activities.

Moreover, changing emission sources are increasing certain forms of AD-N relative to others. For example, intensive animal operations in Western Europe and the U.S. mid-Atlantic region have been linked to elevated ammonium deposition rates (10). In the United States, depositional changes are found in long-term data from the network of National Atmospheric Deposition Program (NADP) sites. The Midwestern and mid-Atlantic states, in particular, have experienced rapid increases in swine and poultry operations since the mid-1980s and have shown a precipitous increase in atmospherically deposited ammonium.

Case study

This trend is illustrated by a coastal NADP site (NC-35) located in Sampson County, North Carolina, which has experienced a large increase in swine operations since the late 1980s (Figure 2). Ammonia volatilization from these intensive animal operations transports N and unintentionally fertilizes distant areas, possibly to their detriment. The fate of this N, up to 90% of which originates in animal feed from grain-producing areas outside of the state, is of particular concern because of the sensitivity of the lower river basins and estuaries in eastern North Carolina to N overenrichment (2).

There is also concern about the amounts and composition of AD-N inputs. Increases in the ratios of ammonium to nitrate in the DON component of AD-N can usher in changes in microbial community composition, because phytoplankton and bacteria may use N forms differently. These changes can translate into other water quality impacts such as undesirable algal blooms. In addition, relative enrichment of N alters the stoichiometric ratio of other essential nutrients like phosphorus and silicon. Iron and trace metal enrichment from AD may synergistically interact with N to stimulate coastal algal production and blooms (11).

Nutrient addition bioassays help assess phytoplankton responses at the functional group leveldiatoms, dinoflagellates, cyanobacteria, cryptomonads, and chlorophytes-to different forms of N. For example, bioassays of water from the N-sensitive Neuse River estuary in North Carolina were amended with equimolar amounts of either ammonium, nitrate, urea, or a combination of the three. Then, the bioassays were either incubated at ambient irradiance or shaded to 10% of surface light levels. Highperformance liquid chromatography-based diagnostic photopigment analyses were used to characterize phytoplankton community responses according to the relative abundance of functional groups (12, 13). Figure 3 shows that different chemical forms of N shifted the community composition at both 100% and 10% of ambient irradiance, which reflect the range of natural light conditions in the estuary. Such shifts may be involved in a recently observed increase in the frequencies and magnitudes of harmful algal blooms in estuarine and coastal waters worldwide. The mechanistic links between shifting amounts and relative proportions of N forms enriching these waters require further investigation.

Research and management perspectives

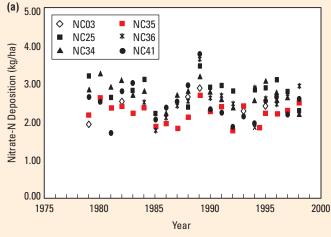
Because it is a major N source, AD-N should be factored into N budgets and considered in maintaining water quality. To do so, we must identify key sources, transformations, transport routes, depositional patterns, and amounts. Because atmospheric emission and deposition patterns are often far greater in scale than watershed boundaries, quantifying AD-N in estuarine N budgets will require both local and regional approaches. This will necessitate airshed-level modeling that is capable of overlapping, in time and space, with watershed and basin-scale hydrologic and nutrient monitoring networks and models.

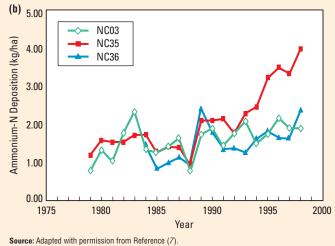
With these needs in mind, research and management questions emerge. What is the contribution of AD-N relative to other new and regenerated N sources in N-sensitive water bodies? How does this vary within and among geographic regions? What is the relative importance of indirect, watershed-mediated versus direct AD-N on estuarine and coastal N budgets, production, and water quality responses? What are the

FIGURE 2

A 20-year record of deposition in North Carolina

(a) Atmospheric nitrate and (b) ammonium wet deposition data were provided by a network of National Atmospheric Deposition Program sites in North Carolina. NC-35 is located in Sampson County, N.C., a region of intensive swine and poultry operations.





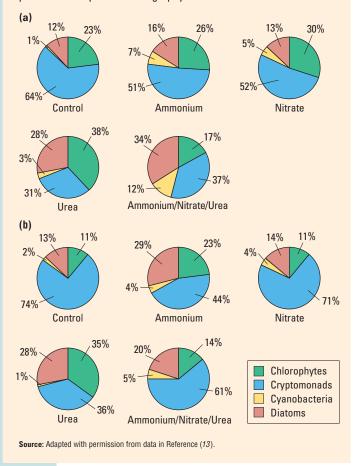
biogeochemical and trophic importance and roles of AD-N for N-sensitive estuaries and coastal and open ocean waters? What are ramifications of specific inorganic and organic AD-N constituents in terms of biogeochemical and trophic responses along this gradient?

There are additional managerial and logistical considerations. Emission sources, routes of transport, deposition, watershed processing, and biological response to atmospheric N need to be spatially and temporally coupled. Appropriately scaled models capable of integrating airshed with watershed N processing are needed. Experiments, monitoring, and

FIGURE 3

Bioassay results

The phytoplankton community responds to different chemical forms of nitrogen administered at 10 micromolar N concentrations. The bioassay was conducted on the Neuse River estuary in North Carolina during spring 1998. Identical sets of samples were incubated at (a) 100% and (b) 10% of surface light levels. Each pie chart represents the total amount of phytoplankton biomass, determined by chlorophyll *a* measurements. This biomass was partitioned into specific phytoplankton functional groups using diagnostic caratenoid photopigments measured by high-performance liquid chromatography.



modeling must be used to evaluate acute versus chronic AD-N biogeochemical and trophic impacts on estuarine, coastal, and open ocean waters. This information must then be incorporated into regional and global assessments of the roles and impacts of AD-N on estuarine and coastal productivity, nutrient cycling, and trophic structure and function. In turn, these assessments must be integrated in local, regional, and national N management schemes and policies aimed at preserving and protecting coastal water quality, habitats, and water resources.

Deciding on the approach

An interdisciplinary monitoring and process-oriented experimental approach should be used to couple AD-N emission sources, transport, and deposition dynamics to biogeochemical and trophic impacts. This should be complemented by modeling efforts aimed at linking AD-N emission, transport, deposi-

tion, and trophic use processes from phytoplankton to fish. Research efforts should be regional in scope and comparative. Emphasis should be placed on comparing sensitivities and impacts of systems exhibiting advanced stages of eutrophication, such as Long Island Sound and the Chesapeake and Delaware Bays, versus those exhibiting incipient stages of eutrophication, such as Tampa Bay, Albemarle–Pamlico Sound, and their adjacent coastal waters.

The roles of AD-N in coastal nutrient dynamics and eutrophication cross land, air, and water interfaces. The interdisciplinary relevance touches on air and water quality research and management, estuarine and marine ecology, environmental modeling, biogeochemistry, microbiology, and basic and applied research looking at the links between atmospheric chemistry and physics and nutrient production. As such, information should be appropriately formatted for formulating policy and making decisions that extend beyond traditional ecosystem and watershed boundaries.

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