Nitrogen Pollution in the Northeastern United States: Sources, Effects, and Management Options

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The northeastern United States receives elevated inputs of anthropogenic nitrogen (N) largely from net imports of food and atmospheric deposition, with lesser inputs from fertilizer, net feed imports, and N fixation associated with leguminous crops. Ecological consequences of elevated N inputs to the Northeast include tropospheric ozone formation, ozone damage to plants, the alteration of forest N cycles, acidification of surface waters, and eutrophication in coastal waters. We used two models, PnET-BGC and WATERSN, to evaluate management strategies for reducing N inputs to forests and estuaries, respectively. Calculations with PnET-BGC suggest that aggressive reductions in N emissions alone will not result in marked improvements in the acid–base status of forest streams. WATERSN calculations showed that management scenarios targeting removal of N by wastewater treatment produce larger reductions in estuarine N loading than scenarios involving reductions in agricultural inputs or atmospheric emissions. Because N pollution involves multiple sources, management strategies targeting all major pollution sources will result in the greatest ecological benefits.

Keywords: atmospheric deposition, nitrogen management, northeastern United States

One of the critical challenges for the sustainable management of natural resources is the accelerating problem of nitrogen (N) pollution. Nitrogen is the most abundant element in the atmosphere as molecular N (N_2). Only after N_2 is converted into reactive forms (Nr; Galloway et al. 2003), such as ammonium (NH_4^+) and nitrate (NO_3^-), is it available to support the growth of plants and microbes. Historically, N fixation by a few specialized organisms accounted for the major inputs of Nr to the biosphere. In the last century, however, human activities have more than doubled the global rate of Nr production (Vitousek et al. 1997) through industrial production of N fertilizers, through atmospheric emissions of Nr associated with fossil fuel combustion, and through cultivation of crops that host microorganisms capable of producing Nr (Smil 2001).

Large changes in the global N cycle have generated concerns that the ecological integrity and environmental health of terrestrial, freshwater, and coastal marine ecosystems are now at risk from oversupply of Nr. Resolution of this problem will require the combined efforts of scientists and policymakers in a way that satisfies food and energy demands while protecting human and ecosystem health.

In this article we examine N pollution from a regional perspective, focusing on the northeastern United States (defined here as New York and New England). The Northeast provides an interesting study region. It receives elevated inputs of Nr, which are highly variable in magnitude and source, reflecting the diverse and rapidly changing landscape. These Nr inputs result in a cascade of environmental effects characterized by interconnected consequences across large spatial scales (Vitousek et al. 1997, Smil 2001, Galloway et al. 2003).

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The Hubbard Brook Research Foundation convened a team of scientists and advisers to answer three questions: (1) What are the inputs of anthropogenic Nr to the northeastern United States? (2) What are the ecological effects of elevated anthropogenic inputs of Nr in the Northeast? (3) What are the management options to mitigate the effects of elevated anthropogenic inputs of Nr? We compiled and analyzed information on the sources and effects of Nr in the northeastern United States. Using two models, we also evaluated various policy strategies to reduce atmospheric N deposition to forest ecosystems and to mitigate the adverse effects of Nr inputs on coastal ecosystems.

Regional and historical context

The population and land cover of the northeastern United States have changed markedly over the last several centuries (figure 1), with important consequences for N retention and export. On their arrival, European settlers cleared forests for crops and pastures. By 1880 approximately 65% of New York and New England had been converted to farmland (figure 1). After the late 1800s, the rise of manufacturing and the westward expansion of agriculture led to farm abandonment and growing urban populations. Supplied by food imported from other parts of the country, the Northeast sustained rapid urban population growth while farmland reverted to forest. By the mid-1990s, farmland had decreased to 17% of the total land area in the Northeast, forest cover had expanded to nearly 75%, and 80% of the population resided in urban areas (figure 1). The second-growth forests that dominate land cover in the Northeast have a large potential capacity to retain deposited N in regrowing trees and reaccumulating soil organic matter (figure 2; Compton and Boone 2000, Goodale et al. 2002).

What are the inputs of anthropogenic reactive nitrogen to the northeastern United States?

Anthropogenically derived Nr reaches the environment through point sources, such as wastewater treatment plant effluent, and nonpoint sources, such as atmospheric deposition and runoff from fertilizer (both chemical and manure) from the landscape.

Emissions and deposition of atmospheric nitrogen. Nitrogen oxides (nitric oxide [NO] and nitrogen dioxide [NO_2], referred to collectively as NO_x) are derived either from the partial oxidation of N_2 at high temperatures or from the release of N contained in fossil fuels during combustion. Major sources of NO_x in the northeastern United States include off-road mobile sources (39%), off-road mobile sources (e.g., logging, pleasure craft, railroads, and lawn and garden equipment; 15%), fossil fuel combustion from electric utilities (25%), and industrial sources (11%) (figure 3; EPA 1998).

Anthropogenic sources of ammonia (NH_3) for the Northeast airshed include agriculture (chemical fertilizers [16%] and animal waste [60%]), human breath and perspiration (7%), domestic animals (7%), sewage treatment plants and septic systems (6%), industrial point sources (2%), and mobile sources (2%) (figure 3; Strader et al. 2001).

Nitrogen oxides and NH_3 can also be transported into the Northeast from emissions sources as far away as the Midwest and mid-Atlantic regions of the United States and portions of southern Canada (figure 3). Canada contributes approximately 13% of the NO_x in the Northeast (figure 3; EPA 1998, Environment Canada 2002).

In addition to inorganic forms of N (NO_x and NH_3), there are several sources of naturally occurring organic N, including sea-spray droplets and plant pollen. Atmospheric organic N may also be derived from anthropogenic inorganic N compounds reacting with non-N-containing organic particles in the atmosphere (Prospero et al. 1996). Organic N typically makes up 30% of atmospheric N deposition (Neff et al. 2002).

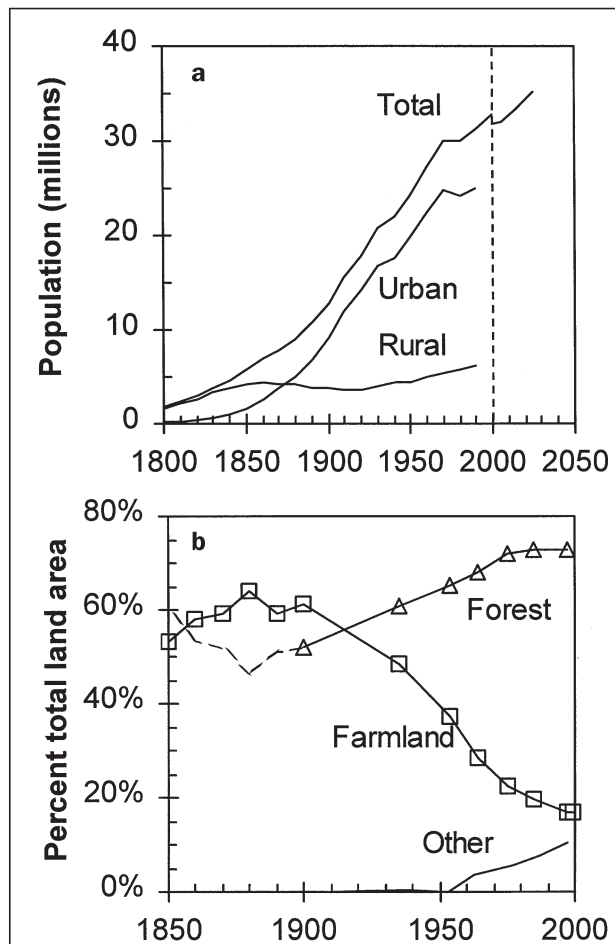

Figure 1. (a) Trends in human population (USCB 1993, 2001) with projections to 2025 (Campbell 1996). (b) Trends in land cover, including forest (Smith et al. 2001), farmland (USCB 1977, USDA 1999), and other lands. Dashed lines indicate estimates. The sum of land area slightly exceeds 100% because data were from different sources and because farmland area often included areas in farm woodlots.

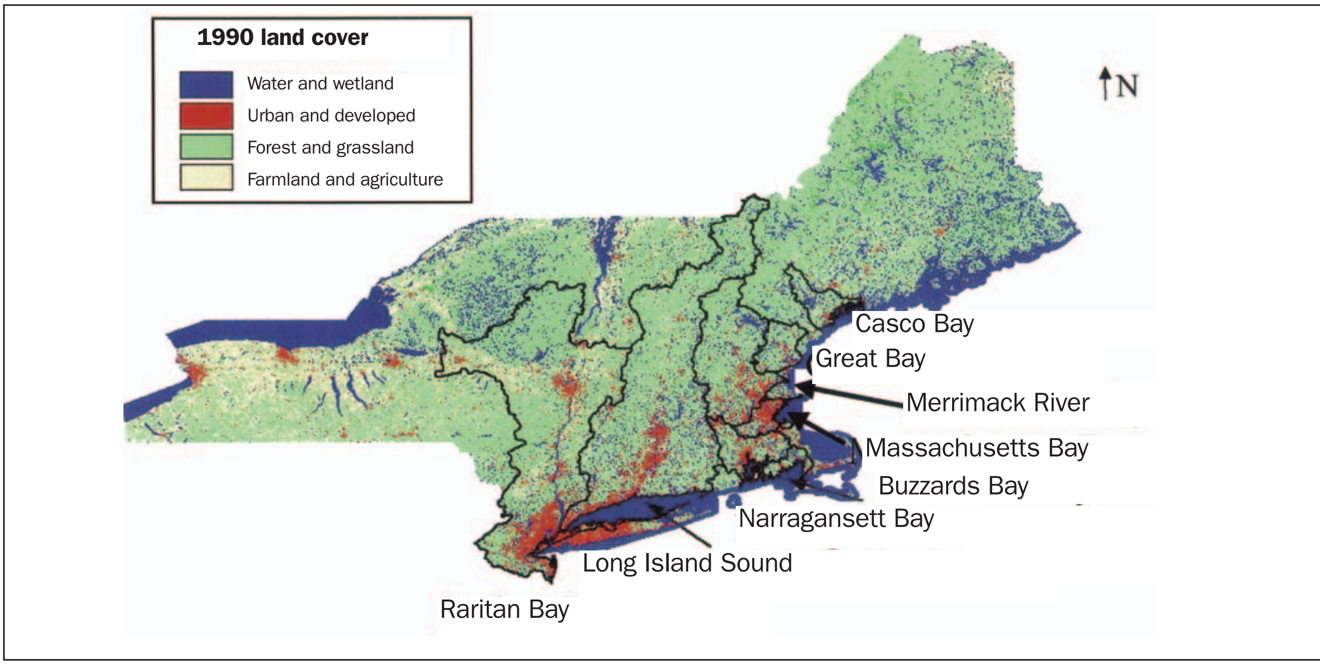

Figure 2. Land cover in 1990 and coastal watershed boundaries. Data from the National Land Cover Database (USGS 1999).

Emissions of N are deposited to Earth in precipitation (wet deposition) and as gases and particles (dry deposition). Wet deposition (rain, snow, sleet, hail, and cloud water) contains a variety of N compounds, most of which are available for biological utilization, including inorganic (NO_3^-, NO_2, NH_4^+) and organic (amino acids, peroxyacetylnitrate, urea) species (Peierls and Paerl 1997). Cloud deposition, which occurs through impaction of fog droplets on exposed surfaces, can contribute between 25% and 50% of total N deposition in high-elevation areas of the Northeast (Anderson et al. 1999). Wet deposition of NO_3^- and NH_4^+ is elevated across the eastern United States and generally decreases from west to east across the Northeast (figure 3). Data from the Hubbard Brook Experimental Forest (HBEF) in central New Hampshire show that concentrations of NO_3^-, NH_4^+, and dissolved inorganic nitrogen (DIN) have been relatively constant in bulk or wet deposition since measurements were initiated in the early 1960s (figure 4). The lack of change in NO_3^- from precipitation is consistent with the relatively constant patterns of NO_x emissions for the Northeast region airshed, even with the enactment of the 1990 Clean Air Act Amendments (CAAA).

Dry deposition includes gaseous compounds or aerosols that are deposited on terrestrial or aquatic surfaces through sedimentation, interception, and diffusion processes. The most prevalent Nr gas species contributing to dry deposition are NH_3 and nitric acid (HNO_3) vapor. Estimates of dry deposition of HNO_3, particulate NH_4^+, and particulate NO_3^- all decrease with increases in latitude (Ollinger et al. 1993).

Nitrogen inputs to watersheds and estuaries in the Northeast. Inputs of Nr to watersheds in the northeastern United States are largely derived from a combination of atmospheric deposition, agricultural activities, and food consumption (Boyer et al. 2002, Castro and Driscoll 2002). The N-rich waste produced by animals (in manure) and humans (in septic systems and sewage) can be an important means of transferring Nr from watersheds to surface waters. This waste comes from point sources (e.g., treated human waste from sewage treatment plants) and from nonpoint sources (e.g., leaching of manure, septic leachate).

To illustrate the patterns of N cycling, we estimated annual net anthropogenic N inputs to eight large watersheds in the Northeast (figure 2). Watershed inputs were calculated for the year 1997 as the sum of five factors: (1) N fertilizer inputs, (2) biotic N fixation by croplands and pasturelands, (3) atmospheric deposition of NH_4^+ and NO_3^-, (4) net import of N in food for humans, and (5) net import of N in feed for livestock (Castro and Driscoll 2002). Input values of total anthropogenic N ranged from 14 kilograms N per hectare per year (kg N per ha per yr) in the Casco Bay watershed, Maine, to 68 kg N per ha per yr in the Massachusetts Bay watershed, Massachusetts (figure 5). In all eight watersheds, net import of N in food for humans was the largest net anthropogenic input, representing 6 to 51 kg N per ha per yr and 38% to 75% of the total (figure 5). Inputs of pet food were not explicitly included in this study but may account for up to 15% of the total N budget for some watersheds (Baker et al. 2001). Atmospheric deposition was the second largest N input for the eight watersheds, ranging from 5 to 10 kg N per ha per yr (11% to 36% of the total). Atmospheric N contributed more than 30% of total anthropogenic N inputs to watersheds of Long Island Sound (35%), Casco Bay (34%), Great Bay (36%), and the Merrimack River (35%). Smaller contributions were attributed to N fertilizer (2 to 13 kg N per ha per yr; 11% to 32% of total N input), N fixation by cropland and pasture-

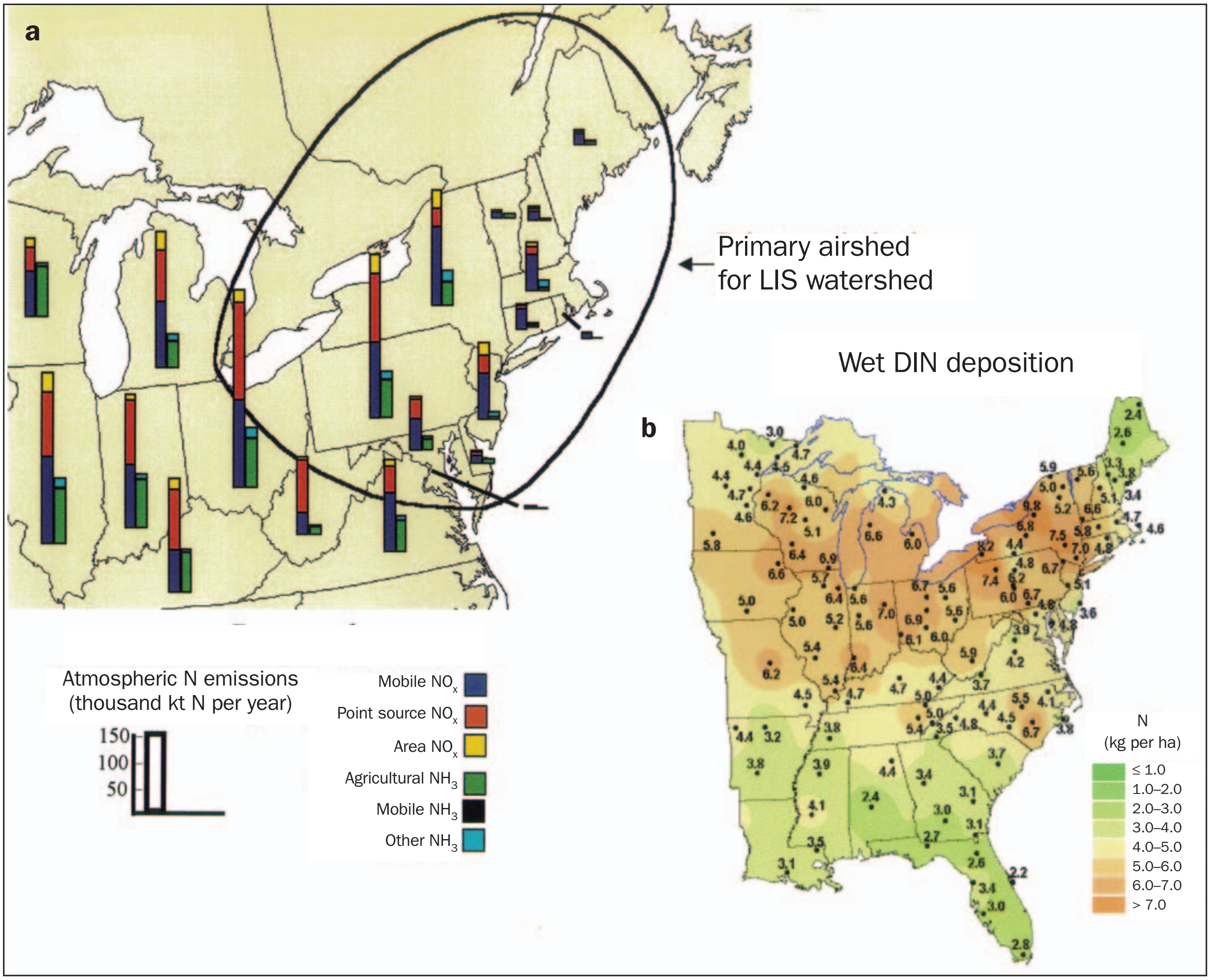

Figure 3. (a) Anthropogenic nitrogen (N) emissions, in thousands of kilotons per year, and (b) wet dissolved inorganic N (DIN) deposition, in kilograms per hectare, for the eastern United States. Nitrogen oxide (NO_x) emissions were obtained from the Environmental Protection Agency for 1996 (EPA 1998); ammonia (NH_3) emissions were obtained from Carnegie Mellon University's Ammonia Emission Inventory for the Continental United States (Strader et al. 2001). The primary airshed is based on regional acid deposition model calculations and shows the area from which the emissions originate that contribute 65% of the deposition to Long Island Sound (LIS; Paerl et al. 2002). Map: National Atmospheric Deposition Program, National Trends Network (NADP 2000).

land (< 1 to 3 kg N per ha per yr; 1% to 8% of total N input), and net feed import of N (< 1 to 3 kg N per ha per yr; 1% to 10% of total N input).

Using the Watershed Assessment Tool for Evaluating Reduction Scenarios for Nitrogen (WATERSN; Castro and Driscoll 2002), we also estimated the contributions of various N sources to the nutrient budgets of estuaries (figure 6) for each of the eight northeastern watersheds. Wastewater effluent was the major source of N loading for all estuaries (36% to 81%). Atmospheric N deposition, either through direct deposition to the estuary surface or through watershed runoff of atmospheric deposition, was generally the second highest source of N (14% to 35%). In addition, runoff from urban areas (< 1% to 20%), agricultural systems (4% to 20%), and forest lands (< 1% to 5%) also contributed N to these coastal ecosystems.

What are the ecological effects of elevated anthropogenic inputs of reactive nitrogen?

The adverse environmental and ecological effects of N pollution result from the contributions of Nr in four major areas: (1) acidic deposition, ground-level ozone (O_3) formation, and visibility loss; (2) acidification and overfertilization of forest ecosystems; (3) acidification and fertilization of fresh waters; and (4) coastal eutrophication. These effects are functionally linked through the N cascade (see Galloway et al. 2003). Effects of atmospheric N emissions on visibility, human health, and materials are beyond the scope of this paper.

Atmospheric effects. Three of the six criteria pollutants for which National Ambient Air Quality Standards (NAAQS) have been established through the Clean Air Act are associated with atmospheric N emissions. Nitrogen dioxide is emitted directly to the atmosphere. Ozone is a secondary pollutant linked indirectly to anthropogenic emissions of NO_x. Particulate matter is partially composed of aerosols containing NO_3^- and NH_4^+ that are formed in the atmosphere following emissions of NO_x and NH_3.

Emission of NO_x can lead to elevated O_3 production through a series of chemical reactions involving volatile organic compounds (VOCs). Because O_3 formation involves both NO_x and VOCs, regulation of O_3 pollution has proved difficult. Early regulatory efforts focused on reducing automobile VOC emissions, which were thought to be the most limiting factor to O_3 production. However, the effectiveness of this strategy has been limited, particularly in humid regions, because of the contribution of biogenic VOCs such as isoprene from vegetation (Chameides et al. 1994). In the eastern United States, it is now recognized that O_3 production is controlled to a greater extent by emissions of NO_x (NRC 1992, Ryerson et al 2001); environmental policymakers have redirected their efforts based on this knowledge.

Many urban and suburban areas of the United States have levels of ground-level O_3 that exceed NAAQS. These areas include a large portion of the Northeast (approximately 95,000 square kilometers [km^2]), with over 26 million people experiencing conditions of elevated O_3 (EPA 2002).

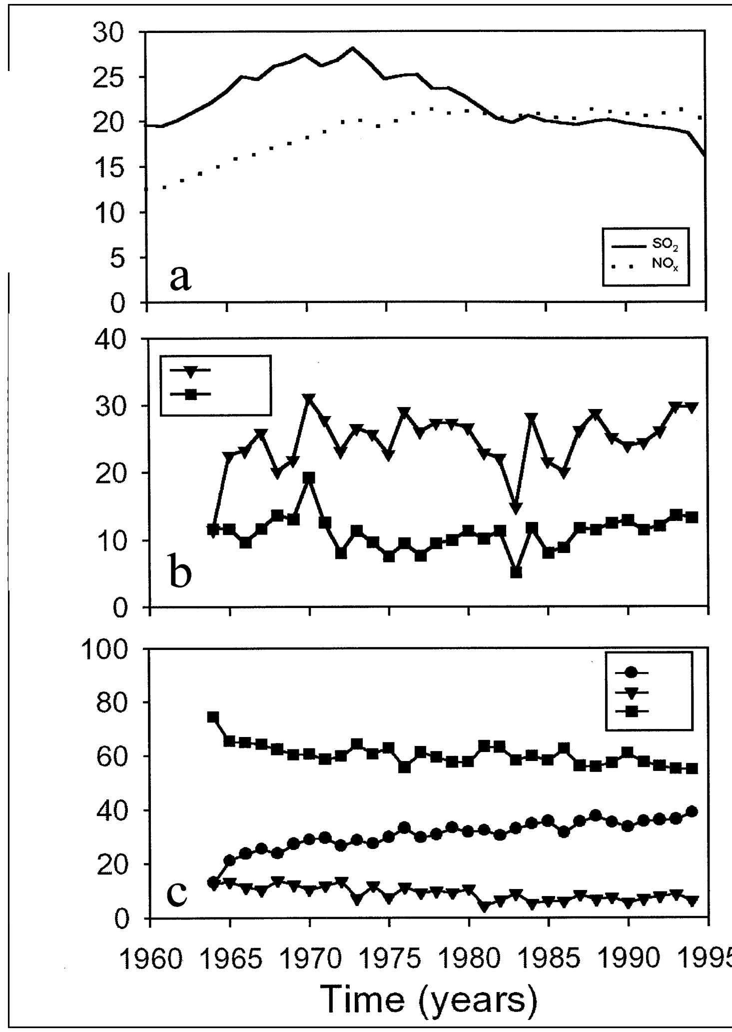

Figure 4. (a) Time series of atmospheric emissions, in megatons (Mt), of sulfur dioxide (SO_2) and nitrogen oxides (NO_x) for the United States; (b) annual volume-weighted bulk deposition, in microequivalents per liter (µeq per L), of ammonium (NH_4^+) and nitrate (NO_3^-); and (c) the equivalent percentage of anions (sulfate [SO_4^{2-}], nitrate [NO_3^-], and chloride [Cl^-]) in bulk deposition at Hubbard Brook Experimental Forest, New Hampshire. Modified from Likens and Lambert 1998.

Terrestrial effects of nitrogen pollution.

Ozone effects on forests and agricultural crops. It has long been recognized that O_3 can have serious negative consequences on the health and function of terrestrial vegetation. The interaction of O_3 with plants occurs primarily through stomatal uptake during periods of active plant growth (Taylor and Hanson 1992). Ozone is a strong oxidant, and injury at the leaf interior is caused by oxidation of cell membranes and photosynthetic enzymes. The most pronounced physiological effect is a reduction in net photosynthetic capacity (e.g., Reich 1987) and associated changes in biomass production and carbon allocation (Laurence et al. 1994). A number of visual symptoms have also been related to foliar O_3 damage (e.g., Gunthardt-Goerg et al. 2000), although their relationship with physiological function is not well established and growth declines are known to occur without any visible sign of injury (Wang et al. 1986).

Variation in sensitivity to O_3 can be caused by a variety of biochemical and morphological factors, although much of the variation observed across species has been related to differences in stomatal conductance (Taylor and Hanson 1992, Kolb et al. 1997). Because conductance is the principal regulator of gas exchange between the leaf and the atmosphere, variation in O_3 sensitivity is caused largely by differences in O_3 uptake. Hence, fast-growing species with high gas exchange rates, including many agricultural crops, tend to be more affected than species with lower inherent growth rates by a given level of O_3 (Reich 1987).

Ozone-related decreases in aboveground forest growth appear to be in the range of 0% to 10% per year (Chappelka and Samuelson 1998). Extrapolation from seedling-level experiments is complicated and uncertain, but process-level modeling offers promise for combining physiological effects with stand- and site-level factors. One such analysis involving the PnET (photosynthesis and evapotranspiration) ecosystem model estimated that O_3 in the northeastern United

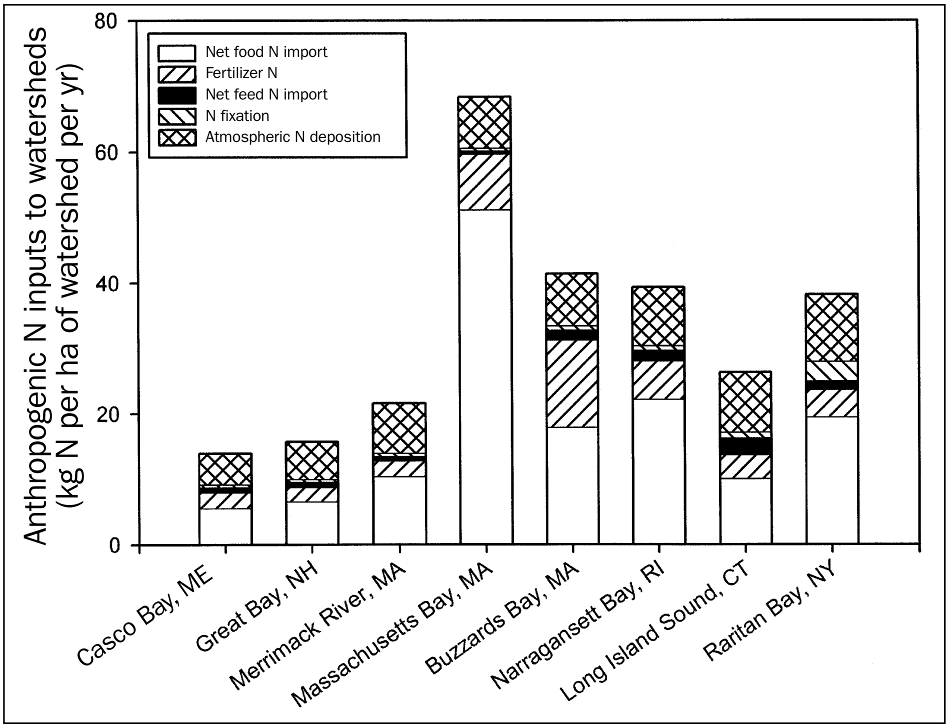

Figure 5. Anthropogenic nitrogen (N) inputs to the watersheds of the northeastern United States, in kilograms per hectare per year.

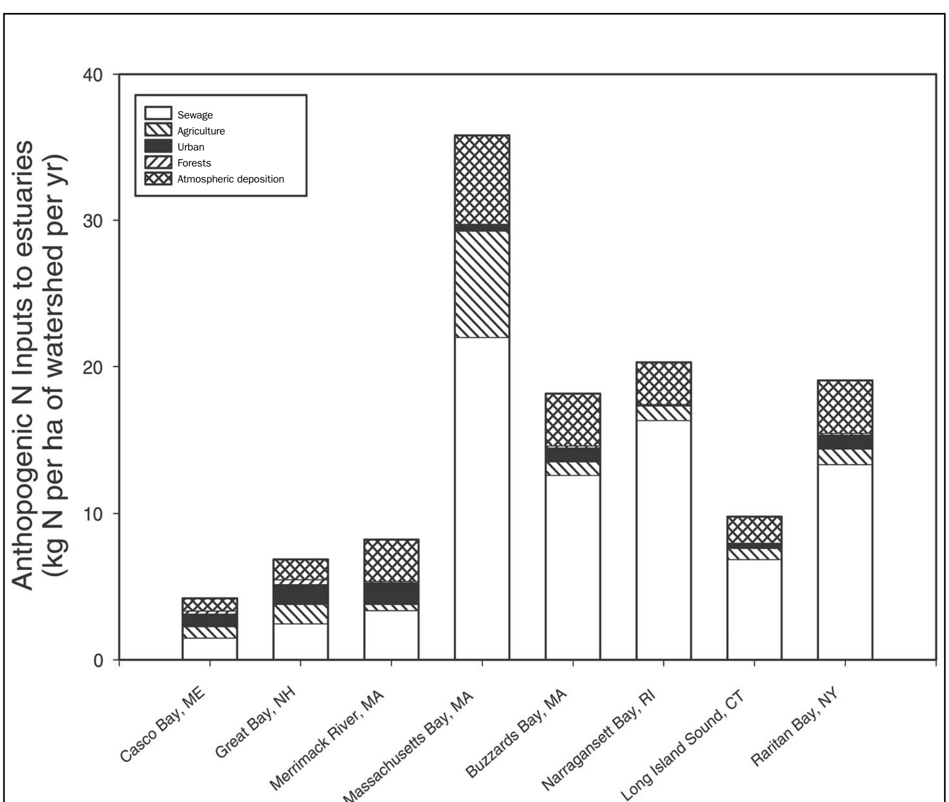

Figure 6. Anthropogenic N inputs to the estuaries of the northeastern United States, in kilograms per hectare per year.

States reduces annual rates of net primary production by 2% to 16%, with variation resulting from differences in O_3 exposure, soil moisture levels, and interactions with other atmospheric pollutants (Ollinger et al. 1997, 2002).

Changes to forest production and nitrogen cycling. Historically, N has been recognized as the nutrient most likely to limit forest growth in temperate and boreal ecosystems. This is at least in part because a long history of extractive land-use practices has reduced N availability and cycling and increased the potential for N retention.

In remote forested watersheds in the Northeast, deposition of atmospheric N is the dominant, and in most cases the single, input of anthropogenic Nr. Cumulative N inputs have fertilized northern forests to the point where some may be at risk from the deleterious effects of Nr oversupply (Nihlgard 1985) or N saturation (Galloway et al. 2003). A number of important changes in forest ecosystem function accompany N saturation, including (a) increased nitrification and NO_3^- leaching, with associated acidification of soils and surface waters; (b) depletion of soil nutrient cations and development of plant nutrient imbalances; and (c) forest decline and changes in species composition. The rate and extent to which these symptoms develop are controlled in part by the capacity of the biota and soils in forest ecosystems to retain deposited N (Aber et al. 1998).

In the northeastern United States, 50% to 100% of atmospheric N inputs are retained by forested watersheds (Aber et al. 2003). Tracer studies using the isotope ^{15}N show that the fraction of N retained in tree biomass ranges from less than 5% in N-poor stands receiving low N deposition to as much as 33% in N-saturated stands receiving high N deposition (Tietema et al. 1998, Nadelhoffer et al. 1999). Much of the remaining N is immobilized by microbial and abiotic soil processes and stored in soil organic matter pools (Bormann et al. 1977, Johnson et al. 2000, Dail et al. 2001). Retention of added N in forest soils should decrease ratios of carbon to nitrogen (C:N) over time, leading to increased rates of net nitrification and increased potential for NO_3^- export. However, the soil C:N ratio is also affected by previous land use history (Compton and Boone 2000, Goodale and Aber 2001), which strongly preconditions the response of forests to N deposition (Aber et al. 1998).

Two of the primary indicators of N enrichment in forest watersheds are the leaching of NO_3^- in soil drainage water and the export of NO_3^- in streamwater, especially during the growing season (Stoddard 1994). These symptoms have been described for watersheds across the Northeast (Aber et al. 2003), including the Adirondack and Catskill Mountains in New York (Cronan 1985, Murdoch and Stoddard 1993, Lovett et al. 2000), the White Mountains in New Hampshire (Goodale et al. 2000), and Bear Brook in Maine (Kahl et al. 1999).

Experimental studies have shown that these symptoms can be induced by chronic additions of N. Ammonium sulfate fertilization of a forest watershed at Bear Brook, Maine, resulted in long-term increases of NO_3^- in streamwater and high annual exports of NO_3^- (Norton et al. 1999). At Harvard Forest, Massachusetts, Magill and colleagues (2000) observed that NO_3^- leaching losses increased continuously over 9 years in treated pine stands but became significant only after 8 years of chronic N additions in an adjacent hardwood stand. Conversely, several N-exclusion studies in Europe demonstrated that decreases in N deposition produced immediate reductions in NO_3^- losses from forest stands (Gundersen et al. 1998, Quist et al. 1999).

While forest growth responses to added N are expected to be positive during early stages, certain forests dominated by evergreen species have shown growth inhibition with chronically elevated N additions (Tamm et al. 1995, McNulty et al. 1996, Magill et al. 2000). On Mount Ascutney, Vermont, additions of less than 31 kg N per ha per yr increased mortality of red spruce *(Picea rubens;* McNulty et al. 1996). The severity of spruce dieback across high-elevation forests in New England in the 1980s correlated with estimated N deposition rates (McNulty et al. 1991). Aber and colleagues (1998) suggest that stands of needle-leaved evergreen forests were more susceptible to growth reductions than broad-leaved deciduous forests.

Chronic inputs of HNO_3 in acidic deposition can accelerate natural processes of soil acidification and increase rates of nutrient cation leaching from the soil profile (Lawrence et al. 1999). Soil acidification may be further enhanced as declining C:N ratios promote increased nitrification rates, resulting in additional proton production. As NO_3^- concentrations increase in acidic northeastern forest soils, there is greater potential for mobilization of aluminum (Al) from soil and interference with divalent cation uptake and root growth by plants (Cronan and Grigal 1995). In addition, elevated concentrations of NH_4^+ from atmospheric deposition can interfere with magnesium uptake and accumulation (Huettl 1990). Lower cation concentrations and lower cation-to-N ratios in foliage have been reported in naturally occurring and experimentally induced N-saturated forests (Aber et al. 1995).

Nitrogen additions to forests can also affect soil microbial processes that control the production and consumption of trace gases, such as nitrous oxide (N_2O), NO, and methane (CH_4), which can affect atmospheric chemistry and global climate. Measurements of N gas effluxes have generally shown small responses to N additions (e.g., Butterbach-Bahl et al. 1997, Magill et al. 1997), although fluxes may be much higher in areas with seasonally elevated water tables (Tietema et al. 1991). To date, fluxes have been measured generally as N_2O only, and losses of NO and N_2 have not been widely examined (Brumme et al. 1999, Groffman et al. 2000). Yet some studies indicate that NO fluxes appear to be more responsive than N_2O to N deposition (Butterbach-Bahl et al. 1997). Soil fluxes of NO contribute to formation of ground-level O_3, and N_2O is a powerful greenhouse gas, while N_2 fluxes remove N from the ecosystem with no negative atmospheric effects (Galloway et al. 2003). Methane is an important greenhouse gas that can be oxidized by microbial processes in aerobic soils. Rates of soil CH_4 consumption are sensitive to N inputs and can be reduced significantly by inorganic N additions (Steudler et al. 1989).

Effects on fresh waters. Atmospheric N deposition can contribute to the acidification of surface waters that drain sensitive upland forest watersheds with limited acid neutralizing capacity (ANC). In contrast, the base-rich soils found in agricultural, suburban, and urban watersheds generally provide sufficient pH buffering to prevent acidification. In the Northeast, surface water acidification resulting from HNO_3 has been characterized as a seasonal and episodic phenomenon associated with high streamflows, in contrast to the chronic acidification associated with sulfuric acid (SO_4^{2-}). More than 30% of the lakes in the Adirondacks and at least 10% of the lakes in New England are susceptible to acidic episodes (Driscoll et al. 2001). Acidic episodes can occur at any time of the year but typically are most severe during spring snowmelt, when biological demand for N is low and saturated soils exhibit lower N retention.

In addition to causing pronounced decreases in pH, acidic episodes induced by NO_3^- also mobilize soil inorganic monomeric Al, which is toxic to fish. For example, brook trout *(Salvelinus fontinalis)*, an acid-tolerant species, is sensitive to concentrations of Al above 3.7 micromoles per liter (µmol per L) (MacAvoy and Bulger 1995). Research has documented the absence of acid-sensitive fish species and the lower density of acid-tolerant fish species in episodically acidic streams (Baker et al. 1996). Effects of acidic episodes include long-term increases in mortality, emigration, and reproductive failure of fish, as well as short-term acute effects. These effects on aquatic life occur despite retention of most atmospheric N deposition within the terrestrial environment. For example,

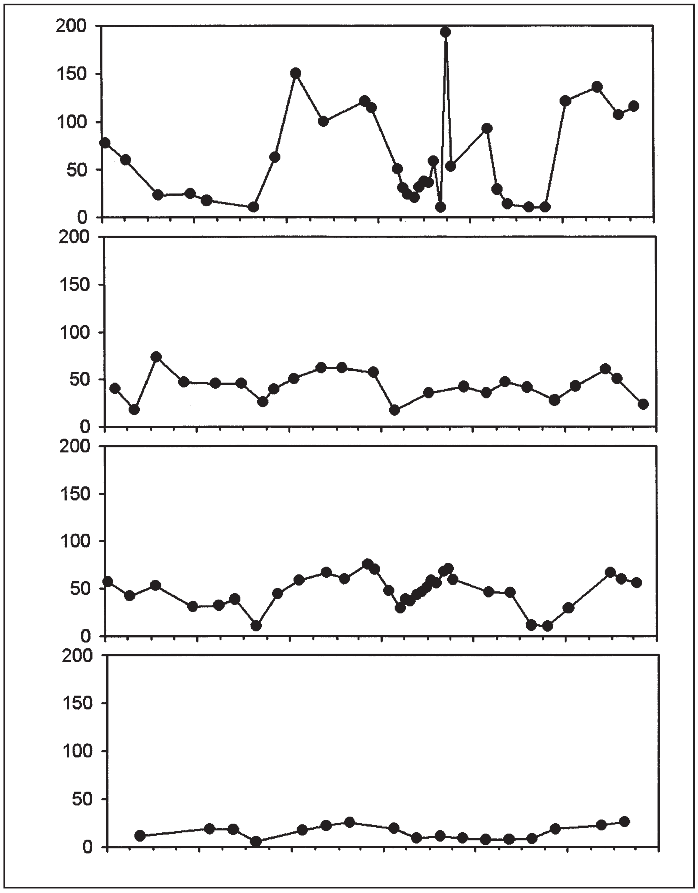

Figure 7. Total dissolved nitrogen (N; the sum of ammonium, nitrate, and organic nitrogen) concentrations, in micromoles per liter, in streams draining watersheds with a predominant land use. (a) Canajoharie Creek, Canajoharie, New York (intensive row crop production); (b) Fall Kill River, Poughkeepsie, New York (urban, sewered); and (c) Lisha Kill Creek, Niskayuna, New York (suburban, residential) are in the Hudson River drainage. (d) Winnisook Creek, near Frost Valley, New York (undisturbed forest), is in the Delaware River drainage, although the sampling site was within 1 kilometer of the Hudson–Delaware divide. All data points represent individual samples except the data from Winnisook Creek, which represent monthly means of approximately weekly sampling.

although 70% to 88% of atmospheric N deposition was retained in a Catskills watershed, fish populations could not be sustained because high NO_3^- concentrations during high flows caused the concentrations of Al to exceed the toxicity threshold (Lawrence et al. 1999).

Although atmospheric sulfur (S) deposition is generally responsible for chronic acidification, Lovett and colleagues (2000) found that NO_3^- concentrations were 37% of SO_4^{2-} concentrations (on an equivalence basis) under baseflow conditions in 39 Catskill streams. These percentages are caused in part by an ongoing decline in SO_4^{2-} concentrations associated with controls on sulfur dioxide (SO_2) emissions, but they also reflect loss of NO_3^- from watersheds throughout the year. On the basis of the classification system of Stoddard (1994), most of these watersheds could be considered to be in the early to middle stages of N saturation.

In the northeastern United States, watershed export of N increases with atmospheric deposition (Aber et al. 2003), particularly above 7 to 8 kg N per ha per yr. Factors such as vegetation type and land-use history affect terrestrial N cycling (Lovett et al. 2000) and contribute to the heterogeneous patterns of watershed N loss in response to variation in atmospheric deposition across the region. Year-to-year variation in N retention may be related to climatic factors that affect microbial dynamics (Murdoch et al. 1998). Using the PnET-CN (carbon and nitrogen) model, Aber and Driscoll (1997) found that climatic factors could explain many of the long-term patterns in stream NO_3^- flux from the biogeochemical reference watershed of HBEF. This complex interaction of factors that affect N biogeochemistry makes it difficult to predict future trends in stream NO_3^- concentrations in forest watersheds.

Concentrations of N in streams of upland forested watersheds tend to be considerably lower than in streams draining watersheds with other land-use characteristics (figure 7). In a comparison of small watersheds in eastern New York, concentrations of N were highest and most variable in a stream draining a watershed where the predominant land use was row crop production. Total dissolved N concentrations in streams in sewered suburban and urban watersheds were somewhat lower and less variable than in the stream draining the agricultural watershed. Streams in urban and suburban watersheds may also experience high episodic N loading caused by combined sewer overflows (CSOs). This source of N is not well quantified, but it may be an important component of N fluxes in urban watersheds under high flow conditions. Fortunately, the US drinking water standard (10 milligrams per L, 714 µmol per L) established to protect infants from methemoglobinemia is rarely exceeded in surface waters and groundwaters in the Northeast (Mueller and Helsel 1996).

At the regional scale, transport of N from terrestrial to freshwater systems has important implications beyond the acidification of upland lakes and streams, because N exports can ultimately contribute to the eutrophication of coastal ecosystems.

Nutrient enrichment and eutrophication in coastal systems. Estuaries and coastal zones are among the most productive ecosystems on Earth (Odum 1971). Nutrient overenrichment is an important stress on many coastal ecosystems of the United States, including areas in New England and New York. In severe cases, it can lead to the development of eutrophic conditions. Nitrogen is the most critical element in coastal ecosystems (Ryther and Dunstan 1971, Oviatt et al. 1995), in contrast with freshwater ecosystems, where primary production and eutrophication are caused largely by excess inputs of phosphorus (Vollenweider 1976).

Coastal eutrophication can cause excessive production of algal biomass, blooms of harmful or toxic algal species, loss

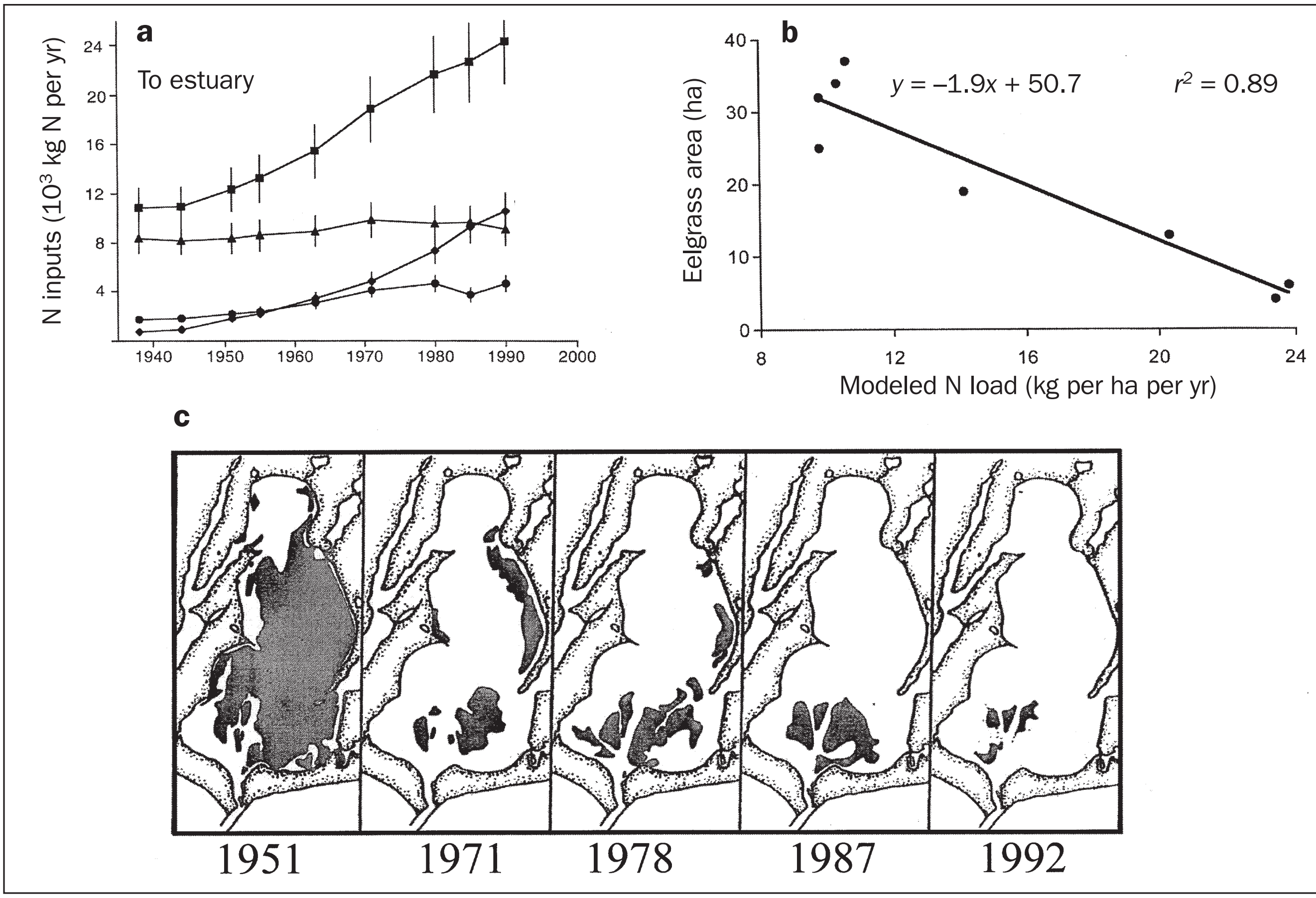

Figure 8. Time course of nitrogen (N) loading and eelgrass coverage in Waquoit Bay between 1938 and 1992 (data from Valiela et al. 2000 and Bowen and Valiela 2001). (a) Modeled historical N loads (in thousands of kilograms per year) to the Waquoit Bay estuary from atmospheric deposition, human wastewater, and fertilizer application. (b) The relationship between N loading (in kilograms per hectare per year) and eelgrass loss (in hectares) as determined from sampling subestuaries within the Waquoit system. (c) Areal extent of eelgrass beds in Waquoit Bay from 1951 to 1992.

of important estuarine habitat such as sea grass beds (figure 8), changes in marine biodiversity and species composition, increases in sedimentation of organic particles, and depletion of dissolved oxygen (hypoxia and anoxia). These primary responses can cause adverse secondary impacts further up the food web (e.g., effects of hypoxia on fish). There are few data documenting the long-term response of coastal ecosystems to changes in N loading. One such data set is available at Waquoit Bay, Massachusetts; it documents increases in N loading to the estuary and the subsequent loss of eelgrass habitat over time (figure 8).

Some estuaries along the Gulf of Maine experience blooms of toxic or harmful algae, frequently called red tides (Anderson 1999). Harmful algal blooms disrupt coastal ecosystems through production of toxins and through the effects of accumulated biomass on co-occurring organisms and food web dynamics. The frequency and geographic extent of harmful algal blooms have increased in recent years; it is hypothesized that this change may be caused by increased nutrient loading (Hallegraeff 1993, Anderson 1995).

Several federal agencies and state, regional, and local organizations recently reported on the status of coastal ecosystems in the United States (Bricker et al. 1999, Summers 2001, SNE 2002). Unfortunately, comprehensive and nationally consistent data on overenrichment are not available for all coastal regions of the United States or for all estuaries in specific regions. However, the National Estuarine Eutrophication Assessment (Bricker et al. 1999) reported that eutrophication was severe in 40% of the total estuarine surface area assessed, including 138 estuaries along the Atlantic, Gulf, and Pacific coasts. Of 23 estuaries examined in the Northeast, 61% were classified as moderately to severely degraded (Bricker et al. 1999).

Nutrient loading is regarded as one of the important drivers of coastal eutrophication. Long Island Sound is a prime example of a northeastern estuary that has experienced eutrophication and hypoxia as a result of long-term N enrichment (NYDEC and CTDEP 2000).

We have a limited understanding of the rate and extent to which estuaries will recover if nutrient inputs from the watershed and the atmosphere are decreased. It is likely that phytoplankton-dominated systems with short hydraulic residence time will reverse their eutrophication trajectories most readily. In contrast, it is likely that benthic-dominated systems with rooted, submerged aquatic vegetation will exhibit delayed recovery.

One example of an ambitious engineering project to mitigate nutrient loading is under way in Boston Harbor. Beginning in 2000, the Massachusetts Water Resources Authority stopped the discharge of wastewater effluent from the Deer Island sewerage treatment plant into Boston Harbor, instead pumping the effluent 15 km offshore into Massachusetts Bay. This change reduced N loading to the harbor by 32 metric tons (t) per day. As a result, there have been rapid improvements in environmental conditions, including an 83% reduction in NH_4^+ concentrations, a 44% reduction in chlorophyll *a* concentrations, and a 12% improvement in water clarity in the harbor (Taylor 2002), largely because it is a phytoplankton-dominated system with a short hydraulic residence time.

What management options exist for reducing nitrogen inputs?

We examined a series of management options for reducing N inputs to forest watersheds and coastal estuaries using two models, PnET-BGC (a biogeochemical ecosystem model) and WATERSN. Possible Nr management options include conservation, control measures, and ecosystem protection. In this analysis we emphasized national public policies for which we could quantify an associated reduction in Nr inputs. We used PnET-BGC (Gbondo-Tugbawa et al. 2001) to examine the response of soil and surface waters in forest watersheds to controls on atmospheric emissions of NO_x and NH_3; we used WATERSN (Castro and Driscoll 2002) to examine the effects of management of N loading to coastal ecosystems.

Nitrogen inputs to air, land, and water are managed through an array of state and federal policies and programs. We developed and evaluated 10 policy scenarios to reduce N inputs in the northeastern United States, based on actual and proposed public policies (table 1).

Reductions in atmospheric emissions of nitrogen. Using PnET-BGC, we evaluated several policy scenarios intended to reduce atmospheric N emissions and deposition (table 1). We examined the predicted changes in the acid–base chemistry of soils and stream water in two forest watersheds (HBEF, New Hampshire, and Biscuit Brook, New York) in response to potential federal controls on atmospheric N emissions. For the emissions scenarios presented here, we assumed that Canadian sources are reduced to the same extent as US sources.

Fossil fuel electric utilities. The 1990 CAAA strive to achieve a 1.8 million t reduction in NO_x emissions from electric utilities by 2010, compared with emission levels expected in the absence of this legislation. Thus, the first scenario we evaluated was the 1990 CAAA scenario, in which NO_x electric utility emissions reach 4.51 million tons per year by 2010 (Driscoll et al. 2001) and remain constant thereafter. For comparison, utility NO_x emissions in the United States totaled 6.04 million t per year in 1996. In evaluating the effects of the 1990 CAAA, we also considered the Environmental Protection Agency (EPA) rule that requires utilities from the 22-state Northeast region to reduce NO_x emissions contributing to ground-level O_3 through state implementation plans. Model calculations using PnET-BGC suggest that this action will not significantly improve the acid–base status of soils or drainage waters, because reductions will be implemented only during the summer growing season and because the total reduction in NO_x emissions will be relatively small (Gbondo-Tugbawa and Driscoll 2002).

For further electric utility scenarios, we assessed recent congressional proposals for additional NO_x emission reductions from electric utilities, ranging from 56% of 1990 levels to 75% of projected 2010 levels (table 1). We evaluated the impact of these emission control proposals using an aggressive utility scenario that assumes a 75% reduction in utility NO_x emissions beyond levels projected in the 1990 CAAA (reduction to 1.13 million tons) for the year 2010.

Transportation sector. Tailpipe emissions from on- and off-road vehicles constitute the largest portion of NO_x emissions in the source area of the northeastern United States (EPA 1998). In 1999, the EPA enacted Tier 2 motor vehicle emission standards to attain and maintain NAAQS for ground-level O_3 and particulate matter. The Tier 2 standards require that the fleet averages 0.07 grams per mile beginning in model year 2004; these standards are phased in for specific categories of vehicles (table 1). In the Tier 2 scenario, we evaluated the NO_x controls planned for the 1990 CAAA combined with the Tier 2 standards.

In addition to the Tier 2 scenario, we also assessed a more aggressive transportation scenario in which a 90% reduction beyond the Tier 2 levels of NO_x emissions from light-duty gasoline vehicles is achieved by converting the current fleet to very low-emission vehicles (Richard Haeuber, EPA, Washington, DC, personal communication, 2002).

Agricultural emissions. Emissions of reduced N (NH_3 gases and NH_4^+ aerosols) from concentrated animal feeding operations (CAFOs) have increased in certain regions over the past decade (Walker et al. 2000) but are not currently regulated. For the purpose of assessing relative impact, we assumed that NH_3 emissions from agriculture could be reduced by 34% through changes in waste storage and treatment. For our last PnET-BGC scenario, called the combination scenario, we combined the aggressive controls for electric utilities, aggressive transportation controls, and reductions in NH_3 emissions. In all the scenarios described above, atmospheric S deposition (and deposition of other elements and meteorological conditions) was projected to remain constant at the 2010 values based on the requirements of the 1990 CAAA.

Reductions in nitrogen loading to coastal watersheds. Anthropogenic N inputs to estuaries in the northeastern United States are largely associated with food and energy production and consumption (Galloway et al. 2003). We developed 10 scenarios for WATERSN to evaluate how potential controls on point and nonpoint sources and on atmospheric

Table 1. Nitrogen reduction scenarios and their policy basis.

Source of N	Model	Scenario	Description	Policy basis
Atmospheric deposition	PnET	1990 CAAA	Projected 9% decrease beyond prior levels	Title IV of the 1990 CAAA includes provisions to reduce acid rain emissions.
	PnET and WATERSN	Aggressive utility	75% reduction beyond 1990 CAAA levels by 2010	This scenario is based on the high end of additional emissions reductions called for in recent legislative proposals.
	PnET and WATERSN	Tier 2 transportation	1990 CAAA plus reduction in transportation NO_x as follows: heavy-duty diesel = 90%; heavy-duty gas = 54%; automobiles = 74%; and off-road vehicles = 60%	The EPA has implemented vehicle emission standards to promote compliance with the National Ambient Air Quality Standards for ground-level ozone and particulate matter.
	PnET and WATERSN	Aggressive transportation	90% reduction in light-duty vehicle NO_x emissions beyond Tier 2	NO_x emissions in this scenario are equivalent to emissions achieved by converting all vehicles to super-low-emission vehicles.
	PnET	Aggressive transportation and 1990 CAAA	90% reduction in light-duty vehicle NO_x emissions beyond Tier 2 with the 1990 CAAA for utilities	This scenario combines the two transportation policies above.
	PnET	Combination	75% reduction in utility emissions beyond 1990 CAAA, 90% reduction in transportation emissions beyond Tier 2, and 34% reduction in agricultural NH_3 emissions	This scenario combines two aggressive scenarios with a proposal to cover and treat waste from CAFOs.
	PnET and WATERSN	Agricultural emissions	34% reduction in agricultural NH_3 emissions	This scenario covers and treats waste from CAFOs in the watershed.
Point sources	WATERSN	Complete BNR	87% reduction through BNR at all WWTPs in the watershed	The Clean Water Act allows the states and the EPA to set TMDLs on nutrients such as N so water quality standards such as those for dissolved oxygen can be achieved. The TMDLs are achieved through reduced riverine loading of N and decreased atmospheric deposition.
		Limited BNR	87% reduction through BNR at WWTPs in the lower portion of the watershed	
		Aggressive wastewater	87% reduction in all septic systems and WWTPs	
		Offshore pumping	100% elimination of N loading from the lower watershed through offshore pumping	The BNR scenarios presented here assume WWTPs are starting from primary treatment levels.
Nonpoint sources	WATERSN	Agricultural runoff	10% and 33% reduction in edge-of-field N loading	The Clean Water Act requires a National Pollution Discharge and Elimination permit for large animal-feeding operations in some cases.The Farm Bill promotes setting aside land to improve water quality.
Multiple sources	WATERSN	Integrated management	87% reduction through complete BNR, 75% reduction in utility emissions beyond the 1990 CAAA, aggressive transportation reductions, 33% reduction in agricultural runoff	This scenario combines several policies described above.

BNR, biotic nitrogen removal; CAAA, Clean Air Act Amendments; CAFO, concentrated animal feeding operations; EPA, Environmental Protection Agency; NO_x, nitrogen oxide; NH_3, ammonia; PnET, photosynthesis and evaporation ecosystem model; TMDL, total maximum daily load; WATERSN, Watershed Assessment Tool for Evaluating Reduction Scenarios for Nitrogen; WWTP, wastewater treatment plant.

Note: The percent reduction in nitrogen in all scenarios is based on the expected reductions at the point of emission or discharge.

deposition would affect N loading to two estuaries in the northeastern United States, Long Island Sound and Casco Bay.

Water quality standards pertaining to N include limits on NO_3^- in drinking water and limits on NH_3 and NO_2 to protect fisheries. These standards are rarely violated in the northeastern United States. However, the EPA has released a guidance document that calls for states and other jurisdictions to develop, by 2004, numeric standards for nutrients such as N in surface waters. At present, N controls for surface waters depend on regulatory programs that limit N inputs to meet other water quality standards (e.g., dissolved oxygen) and to support designated uses (e.g., aquatic life) protected by the Clean Water Act.

When anthropogenic N inputs cause water quality violations, state regulatory agencies are required to develop an EPA-approved total maximum daily load (TMDL) plan that specifies allowable pollutant loading from all contributing sources under applicable water quality standards. A large-scale TMDL has been developed for Long Island Sound. In 2001, the states of Connecticut and New York adopted a plan to address chronic hypoxia in Long Island Sound by reducing N loading to the estuary by 58.5% from target management areas by 2014 (NYDEC and CTDEP 2000). Understanding watershed N inputs and estuarine fluxes of N can help managers achieve these goals.

Wastewater treatment plants. Secondary wastewater treatment plants (WWTPs) are generally not effective in removing N from wastewater. However, biotic N removal (BNR) through denitrification can be added in combination with traditional primary and secondary treatment, thereby reducing N in waste by up to 67% beyond secondary treatment and up to 87% beyond primary treatment alone (NEIWPCC 1998).

Using WATERSN, we considered four scenarios for reducing N inputs from wastewater: (1) the application of BNR technology to all sewered areas in the watershed (basinwide BNR scenario); (2) the application of BNR only to sewered areas in the lower watershed, where N impacts from waste streams would be greatest (near coastal BNR scenario); (3) an enhancement of septic systems to remove N (septic improvement scenario); and (4) displacing human waste generated in the lower watershed of estuaries by pumping wastewater effluent offshore onto the continental shelf (offshore pumping scenario). The last scenario has been implemented or proposed for several estuaries (e.g., Massachusetts Bay), but the long-term ecological effects of offshore pumping to continental shelf benthic systems have not been quantified.

Agriculture. The major agricultural inputs of N in Long Island Sound are fertilizer and manure-laden runoff from fields. Some large animal feeding operations that discharge to US waters are considered point sources of pollution and must obtain a National Pollution Discharge and Elimination System (NPDES) permit (EPA 2001). The EPA has also proposed new regulations for CAFOs. For the purposes of model calculations, we assumed that the agricultural sector could achieve a relatively aggressive 33% reduction in edge-of-field loading of N through improved fertilizer and manure management (agricultural runoff scenario).

Last, we considered an integrated management scenario that evaluated the additive reductions of basinwide tertiary treatment, aggressive mobile NO_x reduction, 75% utilities NO_x reduction, and 33% edge-of-field agricultural runoff reduction.

Model results

Using the PnET-BGC and WATERSN models, we examined the predicted effects of the management scenarios discussed above on forest and coastal ecosystems.

PnET-BGC results. PnET is a simple, generalized, and well-validated model that estimates forest net productivity, nutrient uptake, and hydrologic balances (Aber and Federer 1992, Aber and Driscoll 1997). The model was recently expanded to simulate soil processes and major element biogeochemistry of forest ecosystems (Gbondo-Tugbawa et al. 2001). Because PnET-BGC is a dynamic model, scenarios were considered over the time intervals for which they might be implemented in the future.

The PnET-BGC model was applied to two forest watersheds that have demonstrated sensitivity to atmospheric deposition of strong acids: (1) watershed 6, the reference watershed of HBEF; and (2) Biscuit Brook in the Catskills region of New York. Watershed 6 is characterized by moderate atmospheric N deposition (8.2 kg N per ha per yr) and losses of NO_3^- in streamwater (mean volume-weighted annual concentrations at 20.3 microequivalents [μeq] per L, ranging from 3.8 to 52.9 μeq per L, for 1964–1992; Likens and Bormann 1995, Aber et al. 2003), whereas atmospheric N deposition (11.2 kg N per ha per yr) and stream NO_3^- concentrations are higher at Biscuit Brook (mean volume-weighted annual concentrations at 25.5 μeq per L, ranging from 15.6 to 53.4 μeq per L, for 1983–1999; Murdoch and Stoddard 1993, Aber et al. 2003). PnET-BGC has been previously applied to these watersheds in other analyses (Gbondo-Tugbawa et al. 2001).

We reconstructed historical information on atmospheric deposition and land disturbance (Gbondo-Tugbawa et al. 2001) to simulate soil and stream chemistry over the interval 1850–2000. Our reconstructions of atmospheric deposition reflect estimated changes in inputs of N, S, and other elements to these forest ecosystems in response to changes in emissions. Model hindcasts over this period show that decreases in exchangeable nutrient cation pools, increases in stream concentrations of NO_3^- and SO_4^{2-}, and decreases in ANC have occurred following increases in atmospheric emission and deposition of N and S. Atmospheric S deposition is largely responsible for the acidification of soil and streamwater at these sites and elsewhere in the Northeast (Driscoll et al. 2001). Atmospheric N inputs have, however, made a secondary contribution to acidification, particularly in acid-sensitive areas of New York (i.e., the Adirondacks and the Catskills; Aber et al. 2003).

Model hindcasts indicate that annual volume-weighted concentrations of NO_3^- in 1900 were 1.3 μeq per L at HBEF and 10 μeq per L at Biscuit Brook; these values peaked at HBEF in 1972 (36 μeq per L) and at Biscuit Brook in 1983 (35 μeq per L). Stream NO_3^- concentrations at both locations have decreased somewhat in recent years. To quantify the extent of acidification caused by atmospheric N deposition, we ran PnET-BGC under a scenario of background atmospheric N deposition (i.e., 10% of current values for NO_3^- and NH_4^+) and compared these values with hindcasts based on our estimates of actual historical N and S deposition. This analysis suggests that long-term inputs of atmospheric N deposition by the year 2000 resulted in increases in annual volume-weighted NO_3^- concentrations of 25 μeq per L for HBEF and 19 μeq per L for Biscuit Brook, and decreases in ANC of 7.5 μeq per L for HBEF and 12 μeq per L for Biscuit Brook, compared with conditions expected if atmospheric N deposition were at background levels.

Since the early 1970s, controls on emissions of SO_2 from electric utilities have resulted in decreased atmospheric S deposition (figure 4) and decreased concentrations of SO_4^{2-} in surface waters. The limited recovery of surface waters in New York in response to controls of SO_2 is caused in part by ongoing watershed losses of NO_3^- (Stoddard et al. 1999). As a result, N has developed a more prominent role in regulating the acid–base chemistry of soil and surface waters in forest ecosystems of the Northeast.

Under the 1990 CAAA scenario, PnET-BGC predicts that stream NO_3^- concentrations in both study watersheds will increase over the next 50 years in response to nearly constant atmospheric N deposition to maturing forest ecosystems. The increased concentrations of NO_3^- result in a continued decrease in ANC in streamwater and are consistent with current analysis of time-series data showing a lack of recovery in surface water ANC for areas in the region (Stoddard et al. 1999). Simulations of the response of forest watersheds to changes in atmospheric NO_3^- deposition associated with controls on NO_x emissions from utility and transportation sources, and with controls on NH_3 emissions, show that these scenarios are likely to diminish stream NO_3^- concentrations (table 2). These decreases in atmospheric N deposition are also projected to arrest the acidification of soil and surface waters. Current estimates for atmospheric N deposition at HBEF and Biscuit Brook are slightly above the threshold (7 kg N per ha per yr) established by Aber and colleagues (2003) for elevated NO_3^- leaching in forest ecosystems in the Northeast. Scenarios that call for controls on atmospheric N emissions decrease N deposition to between 4.1 and 7.1 kg N per ha per yr at HBEF and between 4.2 and 7.1 kg N per ha per yr at Biscuit Brook.

Controls on atmospheric N emissions were more effective in decreasing stream NO_3^- and increasing ANC at Biscuit Brook because this site receives higher rates of atmospheric N deposition and shows higher concentrations of NO_3^- in streamwater than HBEF does (table 2). Programs to control NO_x emissions from larger transportation sources are more effective in decreasing stream NO_3^- and increasing stream ANC than proposed utility controls. However, a combination of controls on atmospheric NO_x and NH_3 emissions results in the largest decreases in stream NO_3^- and associated increases in ANC. These predictions are conservative because atmospheric S deposition is expected to continue to decrease in the future (Driscoll et al. 2001) and should accelerate recovery of soils and surface waters affected by acidic deposition. For this analysis, we assumed that climatic conditions were constant at 2000 values. However, we note that model predictions of variations in stream NO_3^- resulting from climatic variations (Aber and Driscoll 1997) are large in comparison with

Table 2. Model calculations using PnET-BGC to compare changes in annual volume-weighted concentrations of nitrate (ΔNO_3^-), in micromoles per liter (μmol/L), and acid neutralizing capacity (ΔANC), in microequivalents per liter (μeq/L), for various scenarios to control atmospheric emissions of nitrogen for watershed 6 at Hubbard Brook Experimental Forest (HBEF) in New Hampshire and Biscuit Brook in the Catskill region of New York. Model calculations are shown for two years, 2030 and 2050.

	HBEF				Biscuit Brook			
	2030		2050		2030		2050	
Scenario	ΔNO_3^- (μmol/L)	ΔANC (μeq/L)	ΔNO_3^- (μmol/L)	ΔANC (μeq/L)	ΔNO_3^- (μmol/L)	ΔANC (μeq/L)	ΔNO_3^- (μmol/L)	ΔANC (μeq/L)
Utility								
Aggressive utility	3.5	0.7	–3.8	1.0	–4.0	1.2	–4.8	1.9
Transportation								
Tier II	–5.3	0.9	–7.2	1.7	–6.3	1.4	–8.5	3.1
Aggressive transportation	–7.9	1.6	–8.7	2.0	–6.1	1.5	–9.5	3.6
Combination								
Aggressive utility, transportation, and agricultural	–10.3	2.0	–12.8	3.1	–11.9	3.0	–14.4	5.9

predictions of NO_3^- resulting from proposed emission control scenarios.

WATERSN results. In addition to reducing atmospheric deposition to forest watersheds, it is necessary to reduce N loading to estuaries. Here we present model-predicted N loadings to Long Island Sound and Casco Bay based on 10 management scenarios (table 1). These watersheds were selected because they have similar land-use patterns but contrasting size (Casco Bay, 2188 km^2; Long Island Sound, 40,744 km^2), population (Casco Bay, 227,000; Long Island Sound, 7,451,000), and net food imports (Casco Bay, 5.6 kg N per ha per yr; Long Island Sound, 10.1 kg N per ha per yr). We used WATERSN to evaluate the N reduction scenarios discussed above.

Using WATERSN, we estimated the amount of N available for transport to estuaries from agricultural lands (crops, orchards, and pastures), urban areas, and upland forests. The amount of N available for export from agricultural lands to estuaries was estimated as the difference between N inputs and N outputs. Nitrogen inputs for our agricultural budgets included N fertilization, N fixation, livestock waste, and atmospheric deposition of NH_4^+ and NO_3^-. Outputs from agricultural lands included crop, livestock, and poultry harvest; volatilization of NH_3; and denitrification. Nitrogen export from urban areas included effluent from WWTPs (point sources), leachate from septic systems, and nonpoint source runoff from pervious and impervious surfaces in urban areas (Neitsch et al. 2001). Atmospheric deposition of NH_4^+ and NO_3^- and nonsymbiotic N fixation were assumed to be the only N inputs to forests.

We estimated N export from upland forests using a nonlinear regression relationship between wet deposition of NH_4^+ and NO_3^- and streamwater export of DIN (NH_4^+ and NO_3^-), using the results of numerous, forest watershed studies in the United States (Castro et al. 2000). We assumed that the contribution of dissolved organic N to the total N load was 50% of the inorganic N load exported from forests. Rates of in-stream retention of N were based on literature values and calibrated by comparing predicted and measured riverine fluxes. Castro and colleagues (2000) provide a detailed description of the mass balance model calculations. The management scenarios considered would most likely be implemented gradually. However, the WATERSN model is not a dynamic model, and therefore we evaluated these strategies only under steady-state conditions.

For both watersheds, strategies to control N from human wastes are more effective in decreasing N loading to the estuaries than strategies to control other sources (figure 9). For Long Island Sound, basinwide BNR with enhanced septic treatment resulted in the largest reduction in estuarine N loading (57%), followed by basinwide BNR alone (53%) and offshore pumping of wastewater effluent (52%). For Casco Bay, basinwide BNR with enhanced septic treatment produced the largest reduction (39%), followed by offshore pumping (38%) and basinwide BNR alone (24%).

Scenarios involving controls on atmospheric N emissions produce relatively small reductions in N loading to Long Island Sound. This is in part because atmospheric N deposition has multiple sources. Reducing only one source of emissions will not have a large effect on the overall estuary N budget. The aggressive utility scenario results in a 1.2% decrease in N loading. The EPA Tier 2 mobile NO_x emissions standards alone result in a 2.5% reduction, and the aggressive mobile scenario in a 3% reduction. Atmospheric N reductions were predicted to have a greater effect on Casco Bay, ranging from 4% (aggressive utility scenario) to 9% (aggressive mobile scenario). Although scenarios to control atmospheric N emissions do not produce the same magnitude of changes as reductions in human waste, an aggressive mobile source reduction plan, in combination with aggressive controls of NO_x from utilities, would produce important reductions in estuarine loading, especially in Casco Bay (13%).

Changes in animal waste treatment are predicted to result in a 13% reduction in N loading to Long Island Sound and a 15% reduction to Casco Bay. There is a 31% reduction for the more agriculturally intensive upper watershed of Long Island Sound. Scenarios involving reductions in edge-of-field agricultural runoff also result in relatively small decreases in loading to both Long Island Sound and Casco Bay (0.5% to 8.3%).

It appears that an integrated management plan, involving reductions from multiple sources, is necessary to achieve the most effective N reduction. We evaluated an integrated management scenario that combines the basinwide BNR scenario, the aggressive utility scenario, the aggressive transportation scenario, and the agricultural runoff scenario. The integrated management scenario results in a 58% reduction in N loading to Long Island Sound and a 45% reduction in loading to Casco Bay. If further N controls are required, the residual N must be targeted for further reductions. For example, of the remaining N inputs to Long Island Sound, 42% are derived from atmospheric deposition, 35% from human wastes, 12% from urban runoff, 8% from agricultural runoff, and 3% from forest runoff. Urban runoff is often associated with large hydrologic events, which dilute untreated sewage in combined sewer systems and exceed the operating capacity of WWTPs. This runoff may be discharged directly into receiving waters as a CSO. The EPA issued a national CSO policy in 1994, suggesting that CSOs will become managed as point sources under the NPDES program of the Clean Water Act. It is possible that these policy tools could result in decreased N loading from CSOs.

It is also important to consider atmospheric and terrestrial impacts, including tropospheric O_3 production, when evaluating the effectiveness of these scenarios. For example, reductions in atmospheric N emissions are less effective than other strategies for reducing N loading to estuaries in the Northeast; however, reductions in atmospheric N emissions may be critical to mitigate the effects of anthropogenic N

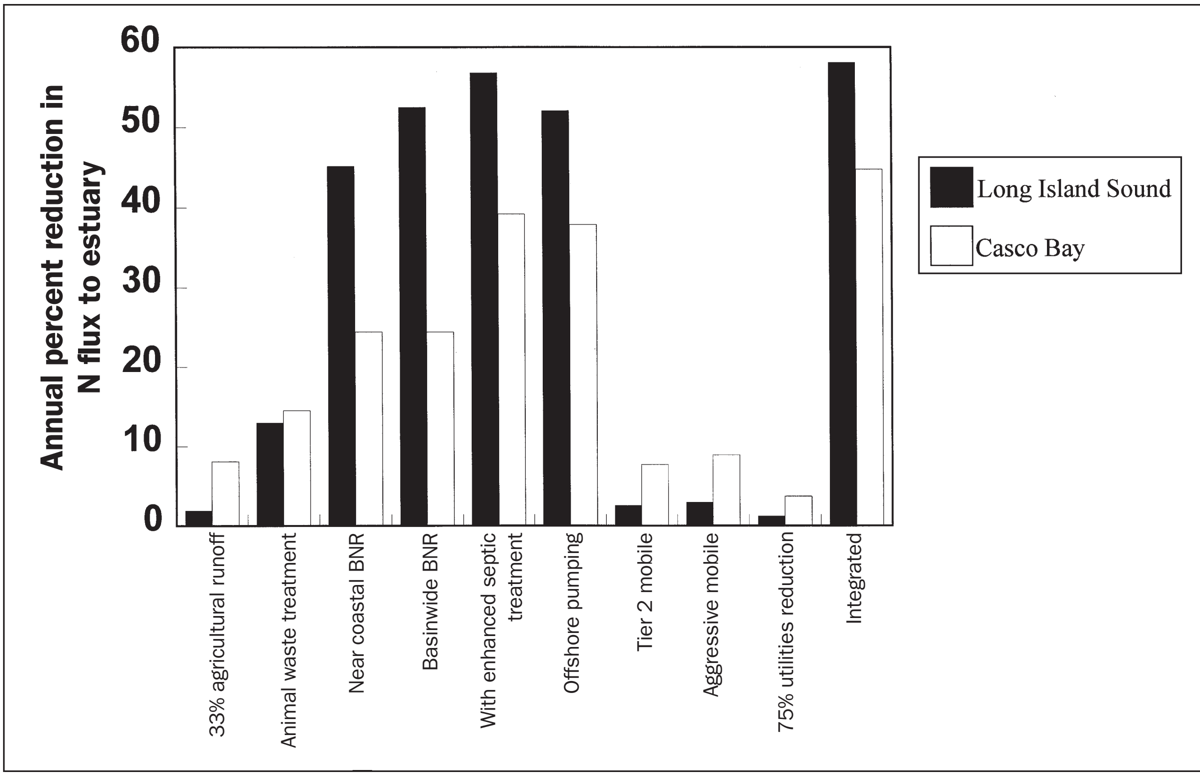

Figure 9. Predicted annual nitrogen (N) flux to Long Island Sound and Casco Bay under various management scenarios using the Watershed Assessment Tool for Evaluating Reduction Scenarios for Nitrogen. BNR is biotic nitrogen removal.

inputs on forest ecosystems, on stream acidification, and on tropospheric O_3 production.

Nitrogen enrichment of natural systems can also be mitigated through strategies targeted toward N conservation or through promotion of landscape characteristics that facilitate the retention of Nr or conversion to N_2 (e.g., wetland protection). However, it is difficult to quantify the ecological effects of such approaches. At the landscape scale, certain ecosystems (e.g., wetlands and riparian zones) can exhibit high rates of N retention and removal, largely through the conversion of Nr to N_2, because of their position at the interface between terrestrial and aquatic environments and because they are characterized by wet, anaerobic soils that support denitrification (Hill 1996). Understanding and managing landscape retention has been identified as a critical component of assessing and controlling N pollution in watersheds (Lowrance et al. 1997, Mitsch et al. 2001, Galloway et al. 2003).

Conclusions

Atmospheric emissions of NO_x and NH_3 are high in the airshed of the northeastern United States, which extends to states in the upper Midwest and mid-Atlantic regions and portions of Canada. Emissions of NO_x are derived largely from electric utilities (36%) and transportation (54%) sources, while NH_3 emissions are derived largely from agricultural (83%) sources. There have not been significant changes in precipitation concentrations of NO_3^- or NH_4^+ since precipitation measurements began at HBEF in the early 1960s. The relatively uniform concentrations and bulk deposition of NO_3^- are consistent with the relatively constant emissions of NO_x for the Northeast in recent decades, despite the 1990 CAAA. In upland forest watersheds like HBEF, atmospheric deposition is the predominant source of N. Together with S, atmospheric N deposition has contributed to the long-term loss of available nutrient cations from soil, the mobilization of elevated concentrations of potentially toxic Al, and the acidification of soil and surface waters. These changes may have adverse effects on terrestrial and aquatic biota.

In contrast to forested uplands, coastal watersheds with urban and suburban lands receive high N inputs from net food imports (38% to 75%). Other sources of anthropogenic N to coastal watersheds of the Northeast include atmospheric N deposition (11% to 36%), N fertilizer inputs (11% to 32%), N fixation by leguminous crops (1% to 8%), and net N feed imports (1% to 10%).

Anthropogenic N inputs are transported to the estuaries of New York and New England by wastewater effluent (36% to 81%), atmospheric deposition to the watershed and estuary (14% to 35%), and runoff from agricultural (4% to 20%), urban (< 1% to 20%), and forest lands (< 1% to 5%). These elevated levels of N loading can result in eutrophication.

Our synthesis shows that sources of N vary across the Northeast landscape. There is a rural-to-urban gradient in which atmospheric deposition contributes N inputs to upland forest watersheds, and wastewater effluent associated with food imports dominates the loading to estuaries.

Model predictions suggest that proposed controls on atmospheric N emissions should decrease NO_3^- concentrations and increase acid-neutralizing capacity in sensitive surface waters draining forest ecosystems; the extent of these changes should coincide with the magnitude of emission controls. The atmospheric N emission control scenarios proposed in this synthesis should decrease atmospheric N deposition to values near or below the threshold (7 kg N per ha per yr) at which elevated NO_3^- leaching occurs for the region. Because atmospheric N deposition is derived from many sources (i.e., utilities, transportation, and agriculture), the most effective mitigation options involve controls on multiple sources of emissions.

This analysis shows that the major source of N to estuaries of the Northeast is wastewater effluent derived mainly from food imports and consumption. Therefore, the most effective single control on N inputs is biotic N removal in wastewater treatment plants. However, in estuaries, as in forest ecosystems, N is supplied from several sources. Therefore, N management strategies to meet anticipated total maximum daily loads should involve controls on major N sources (i.e., wastewater treatment plants, atmospheric deposition, agriculture, and combined sewer overflows) and protection or enhancement of N sinks (e.g., wetlands). Because the response of watersheds to various management scenarios is site specific, it is critical that N issues be assessed on a watershed-by-watershed basis. The rate and extent to which affected forests and estuaries recover from elevated N loading will be an important area of future ecological study as N management plans are implemented.

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