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Effects of acidic deposition on forest and aquatic ecosystems in New York State

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"Capsule": *Elevated inputs of acidic deposition have deleterious effects on forest and aquatic ecosystems in New York.*

Abstract

Acidic deposition is comprised of sulfuric and nitric acids and ammonium derived from atmospheric emissions of sulfur dioxide, nitrogen oxides, and ammonia, respectively. Acidic deposition has altered soil through depletion of labile pools of nutrient cations (i.e. calcium, magnesium), accumulation of sulfur and nitrogen, and the mobilization of elevated concentrations of inorganic monomeric aluminum to soil solutions in acid-sensitive areas. Acidic deposition leaches essential calcium from needles of red spruce, making this species more susceptible to freezing injury. Mortality among sugar maples appears to result from deficiencies of nutrient cations, coupled with other stresses such as insect defoliation or drought. Acidic deposition has impaired surface water quality in the Adirondack and Catskill regions of New York by lowering pH levels, decreasing acid-neutralizing capacity, and increasing aluminum concentrations. Acidification has reduced the diversity and abundance of aquatic species in lakes and streams. There are also linkages between acidic deposition and fish mercury contamination and eutrophication of estuaries.

Keywords: Acidic deposition; Acidification; Soil; Forest ecosystems; Aquatic ecosystems

1. Introduction

Over the past 30 years, scientists have gained greater insight into the ways in which acidic deposition has altered ecosystems. When it was first identified in North America in the early 1970s, acidic deposition was viewed as a simple problem that was limited in scope. Scientists now know that acids and acidifying compounds of atmospheric origin are transported through soil, vegetation, and surface waters, resulting in adverse ecological effects. Further, the same emissions that cause acidic deposition contribute to other important environmental problems, such as visibility degradation, climate change, mercury contamination in fish, and over-fertilization of coastal waters. In this article, we present recent information on trends in acidic deposition, and the ecological effects of acidic deposition, focusing on New York State.

2. Acidic deposition

Acidic deposition is comprised of sulfuric and nitric acids and ammonium derived from emissions of sulfur dioxide, nitrogen oxides, and ammonia. These compounds are largely emitted to the atmosphere by the burning of fossil fuels and by agricultural activities. Once such compounds enter sensitive ecosystems, they can acidify soil and surface waters, bringing about a series of ecological changes. The term acidic deposition encompasses all the forms in which these compounds are transported from the atmosphere to the Earth, including gases, particles, rain, snow, clouds, and fog.

Sulfuric and nitric acids lower the pH of rain, snow, soil, lakes, and streams. pH is a measure of acidity, as determined by the concentration of hydrogen ion. In 1998–2000, wet deposition (i.e., deposition from forms

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of precipitation such as rain, snow, sleet, and hail) in the Adirondack region of New York had an average pH of 4.5 and in the Catskill region had an average pH of 4.4. These values are about 10 times more acidic than background conditions. Wet deposition of sulfate and nitrate is elevated in the Adirondacks and Catskills, and these anions largely contributed to the observed acidity. Wet deposition is highest in the southwestern portion of the Adirondacks and values decrease toward the northeast (Fig. 1; Ito et al., 2002).

Acidic deposition trends in New York and other areas of the Northeast mirror emission trends in the source area which extends to the Midwest (Likens et al., 2000; Butler et al., 2001; Driscoll et al., 2001). Long-term data from the Adirondack and Catskill regions show declining concentrations of sulfate in wet deposition since the late-1970s. Based on these long-term data, it is evident that a strong positive correlation exists between sulfur dioxide emissions in the source area and sulfate concentrations in wet deposition (Fig. 2). Similar relationships have been developed between emissions of sulfur dioxide and wet deposition of sulfate at other sites in New York and New England (Likens et al., 2000; Butler et al., 2001). These observations suggest a cause and effect relationship between emissions of sulfur dioxide and deposition of sulfate in acid-sensitive regions like the Adirondacks and Catskills. It is expected that sulfate concentrations in wet deposition will decrease in a direct linear response to anticipated decreases in sulfur dioxide emissions in the source area. Wet deposition of nitrate has remained relatively constant in New York over the last 20 years, and this observation is consistent with relatively constant nitrogen oxide emissions in the source area for the northeastern USA.

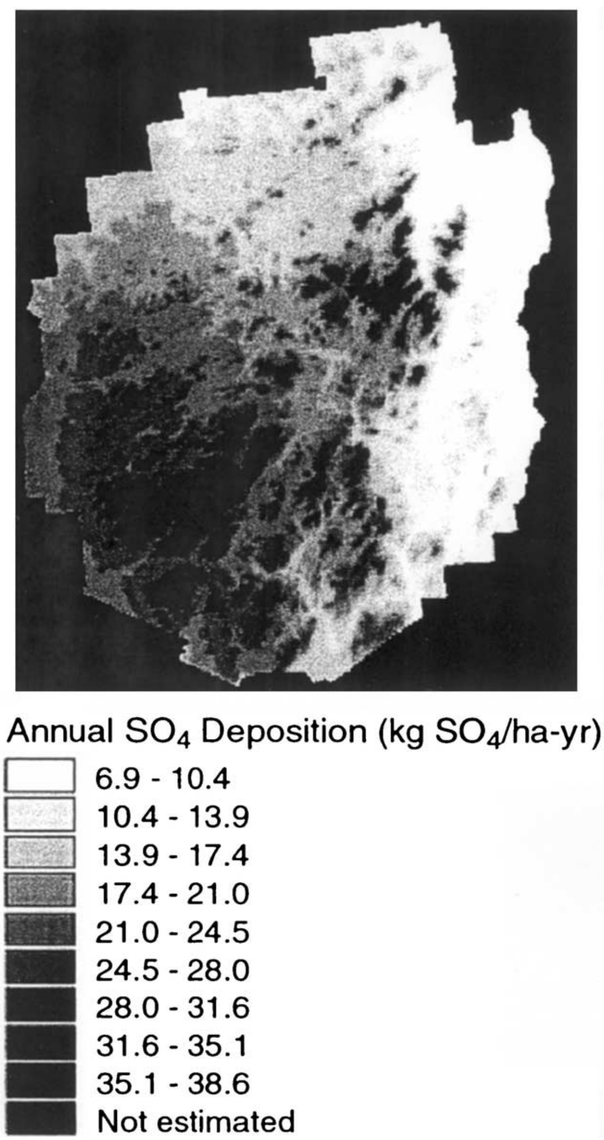

Fig. 1. Spatial patterns of wet sulfate deposition for the Adirondack region of New York (after Ito et al., 2002).

3. Effects of acidic deposition on forest and aquatic ecosystems in New York State

In acid-sensitive regions of New York, like the Adirondacks and Catskills, and elsewhere across Northeast, acidic deposition alters soils, stresses forest vegetation, acidifies lakes and streams, and harms fish and other aquatic life (Driscoll et al., 2001). These effects can interfere with important beneficial uses of ecosystems, such as forest productivity and water quality. Years of acidic deposition have also made many ecosystems more sensitive to continuing strong acid inputs. Moreover, the same pollutants that cause acidic deposition contribute to a wide array of other important environmental issues at local, regional, and global scales (see Table 1).

3.1. Effects of acidic deposition on soils

Recent research has shown that acidic deposition has caused chemical changes in soils in acid-sensitive ecosystems. Soils affected by acidic deposition have diminished ability to neutralize continuing inputs of strong acids, provide poorer growing conditions for vegetation, and extend the time needed for terrestrial and aquatic ecosystems to recover from acidic deposition.

Acidic deposition has altered and continues to alter soils in parts of the Northeast by: (1) depletion of calcium and other nutrient cations from the soil; (2) mobilization of inorganic monomeric aluminum into soil water; and (3) increasing the accumulation of sulfur and nitrogen in soil. Such changes can contribute to adverse impacts on forest health, and the toxicity of surface waters to sensitive biota.

3.1.1. Loss of calcium and other nutrient cations

Over the last century, acidic deposition has accelerated the loss of large amounts of available calcium from

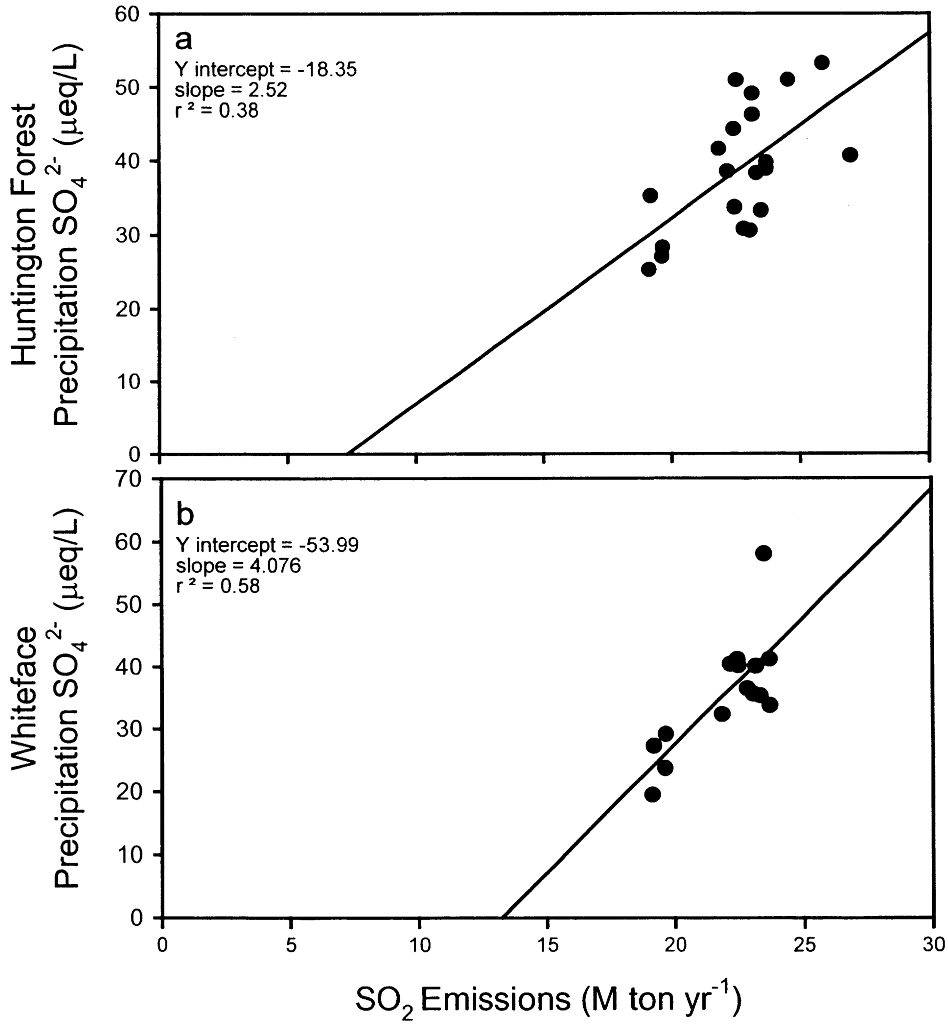

Fig. 2. Relationship between wet deposition of sulfate at the Huntington Forest (a) and Whiteface Mountain (b) in New York and emissions of sulfur dioxide in the source area for the northeastern USA.

Table 1
The links between sulfur dioxide and nitrogen oxide emissions, acidic deposition, and other important environmental issues

Problem	Linkage to acid deposition	Reference
Coastal eutrophication	Atmospheric deposition adds nitrogen to coastal waters.	Jaworski et al., 1997
Mercury	Surface water acidification increases mercury accumulation in fish.	Driscoll et al., 1994
Visibility	Sulfate aerosols diminish visibility and views.	Malm et al., 1994
Climate change	Sulfate aerosols may offset global warming in the short-term, but nitrous oxide is a potent greenhouse gas.	Moore et al., 1997
Tropospheric ozone	Emissions of nitrogen oxides contribute to the formation of ozone with associated adverse effects on vegetation and human health.	NAPAP, 1998

acid-sensitive soil in New York and other acid-sensitive areas in the Northeast (Likens et al., 1996, 1998; Bailey et al., 1996; Lawrence et al., 1999). This conclusion is based on more than a dozen study sites throughout the Northeast, including sites in the Adirondacks, the White Mountains, the Green Mountains, and Maine. Depletion occurs when nutrient cations are displaced from the soil by acidic deposition at a rate faster than they can be replenished by the slow mineral weathering or the deposition of nutrient cations from the atmosphere. This depletion of nutrient cations fundamentally alters soil processes, compromises the nutrition of some trees, and hinders the capacity for sensitive soils and surface waters to recover from acidic deposition.

3.1.2. Mobilization of aluminum

Aluminum is often released from soil to soil water, lakes, and streams in forested regions with high acidic deposition, low stores of available calcium, and high soil acidity (Cronan and Schofield, 1990; Driscoll and Postek, 1995). High concentrations of inorganic monomeric aluminum can be toxic to plants (Cronan and Grigal, 1995), fish (MacAvoy and Bulger, 1995), and other organisms. Concentrations of inorganic monomeric aluminum in streams in New York and New England are often above levels considered toxic to fish (Baker et al., 1990) and much greater than concentrations observed in surface waters draining forest watersheds that receive low inputs of acidic deposition (Driscoll et al., 1988).

3.1.3. Accumulation of sulfur and nitrogen

Acidic deposition results in the accumulation of sulfur and nitrogen in forest soils (Driscoll et al., 2001). As sulfate is released from the soil in response to decreases in the deposition of sulfur, it continues to acidify adjacent streams and lakes. The recovery of surface waters in response to emission controls has therefore been delayed and will not be complete until the sulfur left by a long legacy of acidic deposition is released from soil (Johnson and Mitchell, 1998; Likens et al., 2002).

Similarly, nitrogen has accumulated in soil beyond the amount needed by the forest and appears now to be leaching into surface waters in the Adirondacks (Driscoll et al., 1998) and many other parts of the Northeast. Although forests require nitrogen for growth, several recent studies suggest that in some areas of the Northeast, such as the Catskills and Adirondacks, nitrogen levels are above what forests can use and retain (Stoddard, 1994; Fenn et al., 1998; Mitchell et al., 2001).

3.2. Effects of acidic deposition on trees in areas of the Northeast

Recent research shows that acidic deposition has contributed to the decline of red spruce and sugar maple trees in the eastern USA. Symptoms of tree decline include poor crown condition, reduced tree growth, and unusually high levels of tree mortality. Declines of red spruce and sugar maple in the northeastern USA have occurred during the past four decades. Factors associated with declines of both species have been studied and include important links to acidic deposition (Driscoll et al., 2001).

3.2.1. Red spruce

Since the 1960s, more than half of the large canopy trees in the upper elevations of the Adirondack Mountains of New York and the Green Mountains of Vermont, and approximately one quarter of the large canopy trees in the upper elevations of White Mountains of New Hampshire have died (Craig and Friedland, 1991). Significant growth declines and winter injury to red spruce have been observed throughout its range (DeHayes et al., 1999). Acidic deposition is believed to be a contributing factor in red spruce decline at high elevations in the Northeast. Red spruce decline occurs by both direct and indirect effects of acidic deposition. Direct effects include the leaching of calcium from needles of trees (DeHayes et al., 1999), whereas indirect effects refer to changes in the underlying soil chemistry (Shortle and Smith, 1988; Cronan and Grigal, 1995).

Recent research suggests that the decline of red spruce is linked to the leaching of calcium from cell membranes in spruce needles by acid mist or fog (DeHayes et al., 1999). The loss of calcium renders the needles more susceptible to freezing damage, thereby reducing the tolerance of trees to low temperatures and increasing the occurrence of winter injury and subsequent tree damage or death. In addition, low calcium and elevated aluminum concentrations in the soil may result in a reduction in biomass and limit root uptake of water and nutrients, contributing to decline (Raynal et al., 1990).

3.2.2. Sugar Maple

The decline of sugar maple has been studied in the eastern USA since the 1950s. Extensive mortality among sugar maples appears to have resulted from deficiencies of nutrient cations, coupled with other stresses such as insect defoliation or drought (Drohan et al., 1999; Houston, 1999; Horsley et al., 1999, 2000). Some studies suggest that the probability of the loss of sugar maple crown vigor or incidence of tree death increases on sites where the supply of calcium and magnesium to soil and foliage is low and stress from insect defoliation and/or drought is high (Horsley et al., 1999, 2000). In northwestern and north central Pennsylvania, soils on the ridgetops of unglaciated sites contain low calcium and magnesium as a result of more than half a million years of weathering combined with the leaching of these elements by acidic deposition. Low levels of nutrient cations can cause a nutrient imbalance and reduce the ability of a tree to respond to stresses such as insect infestation and drought.

In addition to the forest stands in Pennsylvania, forests in areas of New York contain sugar maple with foliage having low calcium and magnesium concentrations. Sugar maple in these forests may be susceptible to decline if stress levels surpass a threshold.

3.3. Surface waters

3.3.1. Acidification of surface waters

Acidic deposition degrades water quality by lowering pH levels (i.e. increasing acidity); decreasing acid-neutralizing capacity (ANC); and increasing aluminum concentrations. While sulfate concentrations in lakes and streams have decreased over the last 20 years, they remain high compared to background conditions (e.g.

approximately 20 μeq/l; Driscoll et al., 1988, 1991). Moreover, improvement in other chemical conditions in lakes and streams of the Adirondacks and Catskills has been limited (Stoddard et al., 1999; Driscoll et al., 2001).

A comprehensive survey of lakes greater than 0.2 ha in surface area in the Adirondack region of New York was conducted by the Adirondack Lakes Survey Corporation to obtain detailed information on the acid-base status of waters in this region (Kretser et al., 1989). Of the 1469 lakes surveyed, 24% had summer pH values below 5.0. Also 27% of the lakes surveyed had values of ANC less than 0 μeq/l and an additional 21% had ANC values between 0 and 50 μeq/l. Note that 64% of these acid-sensitive lakes (388 lakes) were characterized by relatively low concentrations of dissolved organic carbon (i.e. $<$7.2 mg C/l; 600 μmol C/l). These lakes had a chemical composition which suggests that their acidity was largely derived from sulfate associated with acidic deposition (Table 2). In contrast, 36% of the lakes (197 lakes) were characterized by high concentrations of dissolved organic carbon (i.e. $>$7.2 mg C/l; 600 μmol C/l) and naturally occurring organic acids (Table 2). These lakes were probably naturally acidic. While the contribution of naturally occurring acidity was greater in these latter lakes, sulfate remains the dominant anion in the solution chemistry and the acidity of these lakes has been clearly increased by acidic deposition.

A survey of the chemistry of stream water under spring base-flow conditions was conducted in the Catskills (and other areas of the eastern USA) in 1986 by the USA Environmental Protection Agency (Kaufman et al., 1988). They estimated that 1–2% of the total stream length (53–108 km) had ANC values $<$50 μeq/l. The length of streams with low ($<$50 μeq/l) and acidic ($<$0 μeq/l) ANC values greatly increases during spring high flow conditions.

Seasonal acidification is the periodic increase in acidity and the corresponding decrease in pH and ANC in streams and lakes (Wigington et al., 1996). Episodic acidification is caused by a sudden pulse of acids and/or a dilution of base cations (e.g. calcium, magnesium, sodium, potassium) due to spring snowmelt and large rain events throughout the year. Increases in nitrate are generally important to the occurrence of acid episodes in the Adirondacks and Catskills, especially when trees are dormant and therefore uptake of nitrogen by vegetation is low. Episodic acidification often coincides with pulsed increases in concentrations of inorganic monomeric aluminum. Short-term increases in acid inputs to surface waters can reach levels that are lethal to fish and other aquatic organisms (Baker et al., 1996; Van Sickle et al., 1996).

Regional trends in surface water chemistry indicate that recovery of sensitive lakes and streams throughout the Northeast from recent decreases in acidic deposition is slow (Stoddard et al., 1999). Twenty-five lakes and streams in the Adirondack and Catskill Mountains and seventeen in New England have been intensively monitored since 1982. A recent analysis shows that these lakes and streams have shown decreases in concentrations of sulfate (Stoddard et al., 1999). This pattern is consistent with decreases in emissions of sulfur dioxide and atmospheric deposition of sulfate. Lakes and streams in the Adirondack and Catskill regions have exhibited limited recovery in pH and ANC, as well as continued acid episodes (Stoddard et al., 1999).

Three factors account for the slow recovery in chemical water quality in the Adirondacks and Catskills, despite the decreased deposition of sulfur associated with the Clean Air Act. First, levels of acid-neutralizing base cations in streams have decreased markedly due to a loss of available base cations from the soil and, to a lesser extent, a reduction in atmospheric inputs of base cations (Likens et al., 1996). Second, atmospheric nitrogen inputs have resulted in elevated concentrations of nitrate in surface waters, contributing to acidification (Stoddard, 1994). Finally, sulfur has accumulated in the soil and is now being released to surface water as sulfate, even though sulfate deposition has decreased (Gbondo-Tugbawa et al., 2002; Likens et al., 2002).

3.3.2. *Response of aquatic biota to acidification of surface waters by acidic deposition*

Decreases in pH and elevated concentrations of aluminum have reduced the species diversity and abun-

Table 2
Concentrations, standard deviation (SD) and% of total anions in acid-sensitive Adirondack lakes (acid neutralizing capacity $<$50 μeq/l) that have low concentrations of dissolved organic carbon (DOC $<$7.2 mg C/l; 600 μmol C/l; 388 lakes) and high concentrations of dissolved organic carbon (DOC $>$7.2 mg C/; 600 μmol C/l; 197 lakes)

	DOC $<$7.2 mg C/l			DOC $>$7.2 mg C/l		
	Conc. (μeq/l)	S.D.	% of total anions	Conc. (μeq/l)	SD	% of total anions
SO_4^{2-}	103.6	27.1	77.9	78.8	27.1	52.8
NO_3^-	0.7	1.3	0.5	0.2	0.5	0.1
Cl	7.2	5.9	5.4	14.6	62.0	9.8
F	2.9	1.6	2.2	3.4	2.0	2.3
HCO_3^-	1.2	2.6	0.9	1.8	2.8	1.2
Organic anions	17.5	24.3	13.2	50.5	32.8	33.9

dance of aquatic life in many streams and lakes in acid-sensitive areas of New York and other areas of the Northeast. Fish have received the most attention to date, but entire food webs are often adversely affected (Schindler et al., 1985).

Decreases in pH and increases in aluminum concentrations have diminished the species diversity and abundance of plankton, invertebrates, and fish in acid-impacted surface waters in the Northeast (Baker et al., 1990; Table 3). In the Adirondacks, a significant positive relationship exists between the pH and ANC levels in lakes and the number of fish species present in those lakes (see Fig. 3; Gallagher and Baker, 1990). Surveys of 1469 Adirondack lakes conducted in 1984 and 1987 show that 24% of lakes (i.e. 346) in this region do not support fish (Gallagher and Baker, 1990). These lakes had consistently lower pH and ANC, and higher concentrations of aluminum than lakes that contained one or more species of fish (Fig. 4). Experimental studies and field observations demonstrate that even acid-tolerant fish species such as brook trout have been eliminated from some waters in New York (Gallagher and Baker, 1990).

Although chronically high acid levels stress aquatic life, acid episodes are particularly harmful because abrupt, large changes in water chemistry allow fish few areas of refuge. High concentrations of aluminum are directly toxic to fish and pulses of aluminum during acid episodes are a primary cause of fish mortality (MacAvoy and Bulger, 1995; Van Sickle et al., 1996; Baker et al., 1996). High acidity and aluminum levels disrupt the salt and water balance of blood in fish, causing red blood cells to rupture and blood viscosity to increase. Studies show that the viscous blood strains the heart, resulting in a lethal heart attack (MacAvoy and Bulger, 1995).

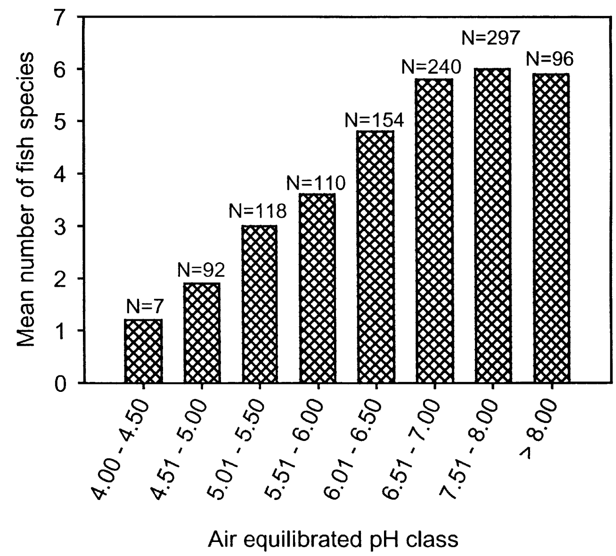

Fig. 3. The mean number of fish species for pH classes from 4.0 to 8.0 in lakes in the Adirondack region of New York. N represents the number of lakes in each pH class (modified from Kretser et al., 1989; Driscoll et al., 2001).

Table 3
Biological effects of surface water acidification (after Baker et al., 1990)

pH decrease	General biological effects
6.5–6.0	Small decrease in species richness of phytoplankton, zooplankton, and benthic invertebrate communities resulting from the loss of a few highly acid-sensitive species, but no measurable change in total community abundance or production. Some adverse effects (decreased reproductive success) may occur for highly acid-sensitive species (e.g., fathead minnow, striped bass)
6.0–5.5	Loss of sensitive species of minnow and dace, such as blacknose dace and fathead minnow; in some waters decreased reproductive success of lake trout and walleye, which are important sport fish species in some areas. Visual accumulations of filamentous green algae in the littoral zone of many lakes, in some streams. Distinct decrease in the species richness and change in species composition of the phytoplankton, zooplankton, and benthic invertebrate communities, although little if any change in total community biomass or production
5.5–5.0	Loss of several important sport fish species, including lake trout, walleye, rainbow trout, and smallmouth bass; as well as additional nongame species such as creek chub. Further increase in the extent and abundance of filamentous green algae in lake littoral areas and streams. Continued shift in the species composition and decline in species richness of the phytoplankton, periphyton, zooplankton, and benthic invertebrate communities; decrease in the total abundance and biomass of benthic invertebrates and zooplankton may occur in some waters. Loss of several additional invertebrate species common in oligotrophic waters, including *Daphnia galeata mendotae, Diaphanosoma leuchtenbergianum, Asplanchna priodonta*; all snails, most species of clams, and many species of mayflies, stoneflies, and other benthic invertebrates. Inhibition of nitrification
5.0–4.5	Loss of most fish species, including most important sport fish species such as brook trout and Atlantic salmon; few fish species able to survive and reproduce below pH 4.5 (e.g., central mudminnow, yellow perch, and in some waters, largemouth bass). Measurable decline in the whole-system rates of decomposition of some forms of organic matter, potentially resulting in decreased rates of nutrient cycling. Substantial decrease in the number of species of zooplankton and benthic invertebrates and further decline in the species richness of the phytoplankton and periphyton communities; measurable decrease in the total community biomass of zooplankton and benthic invertebrates in most waters. Loss of zooplankton species such as *Tropocyclops prasinus mexicanus, Leptodora kindtii,* and *Conochilis unicornis*; and benthic invertebrate species, including all clams and many insects and crustaceans. Reproductive failure of some acid-sensitive species of amphibians such as spotted salamanders Jefferson salamanders, and the leopard frog.

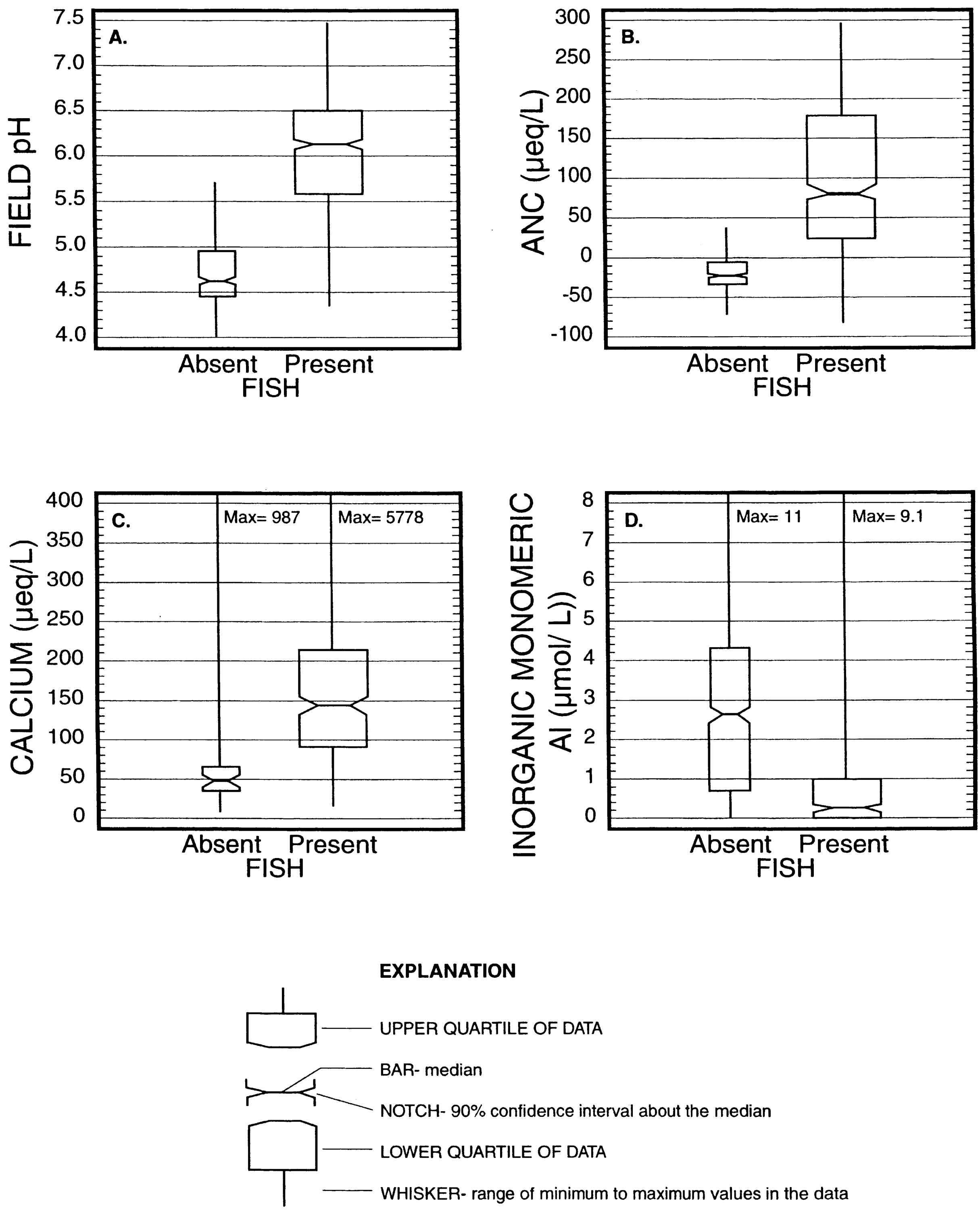

Fig. 4. Chemical characteristics of lakes in the Adirondack region of New York in which fish are absent and present. The chemical characteristics shown include pH, acid neutralizing capacity, calcium concentration and concentrations of inorganic monomeric aluminum.

3.3.3. Linkages between surface water acidification and fish mercury concentrations

Studies in the Adirondack region of New York have shown that many lakes and reservoirs have elevated concentrations of mercury in fish tissue (Driscoll et al., 1994). Mercury contamination of fish is coupled to surface water acidification through the observation of increases in fish mercury concentration with decreases in lake pH (Fig. 5). In a survey of Adirondack lakes, 33% of the yellow perch caught exceeded the 0.5 µg/g Action

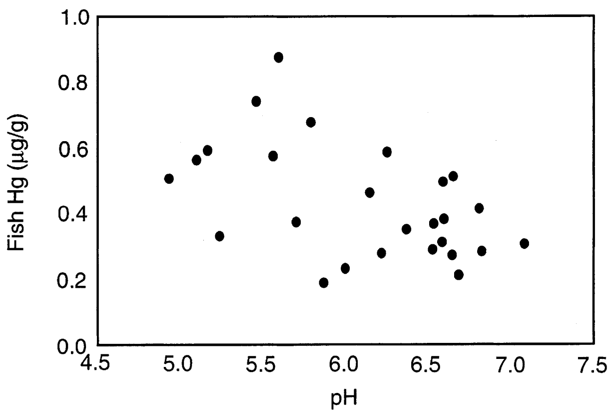

Fig. 5. Concentrations of mercury in yellow perch (year classes 3+ to 5+) as a function of pH for lakes in the Adirondack region of New York (modified after Driscoll et al., 1994).

Level, while 7% exceeded the U.S. Food and Drug Administration Action Level of 1.0 µg/g (Driscoll et al., 1995). Additionally one or more perch with Hg concentrations exceeding the 0.5 µg/g level were found in 88% of the study lakes and one or more perch exceeding the 1.0 µg/g level were found in 56% of the study lakes (Driscoll et al., 1995).

3.3.4. *Atmospheric nitrogen deposition and eutrophication of coastal waters*

Nitrogen is a key nutrient controlling the productivity and eutrophication of estuaries (Ryther and Dunstan, 1971; Nixon, 1986; Fisher and Oppenheimer, 1991; D'Elia et al., 1992). Elevated nitrogen inputs to estuaries leads to increased frequencies of harmful algal blooms, hypoxic and anoxic bottom waters, loss of sea grasses and reduced fish stocks (Valiela and Costa, 1988; Valiela et al., 1990; Hallengraeff, 1993; Boynton et al., 1995; Paerl, 1988, 1995, 1997). These unwanted eutrophication problems are primarily related to human-induced increases in nitrogen inputs to estuaries and are likely to persist or even expand in the future as a consequence or increased population growth in coastal regions and increases in air pollution (Lee and Olsen, 1985; Peierls et al., 1991; Nixon, 1995; Lapointe and Matzie, 1996; Vitousek et al., 1997). A significant portion of nitrogen inputs to estuaries in New York may be derived from atmospheric deposition (Jaworski et al., 1997). For example, Castro et al. (in press) estimate that atmospheric deposition contributes 16% of the total nitrogen loading to both Long Island Sound and Hudson River/Raritan Bay.

4. Summary

New York State and other areas of the northeastern USA receive elevated inputs of acidic deposition. Acidic deposition refers to high loadings of sulfur, nitrogen and acidity to the Earth's surface due to fossil fuel combustion associated with electric utilities, industry and transportation sources, and agricultural activities. In New York, ecological effects of acidic deposition include the acidification of soil and surface waters, decreases in the diversity of aquatic organisms in acid-impacted waters, increased accumulation of mercury in fish tissue, enhanced eutrophication of coastal waters, and increased stress to sugar maple and red spruce.

References

Bailey, S.W., Hornbeck, J.M., Driscoll, C.T., Gaudette, H.E., 1996. Calcium inputs and transport in a base-poor forest ecosystem as interpreted by Sr isotopes. Water Resources Research 32, 707–719.

Baker, J.P., Van Sickle, J., Gagen, C.J., DeWalle Jr, D.R., Sharpe, W.F., Carline, R.F., Baldigo, B.P., Murdoch, P.S., Bath, D.W., Kretser, W.A., Simonin, H.A., Wigington, P.J., 1996. Episodic acidification of small streams in the northeastern United States: effects on fish populations. Ecological Applications 6, 422–437.

Baker, J.P., Bernard, D.P., Christensen, S.W., Sale, M.J., 1990. Biological Effects of Changes in Surface Water Acid-base Chemistry (report SOS/T 13). National Acid Precipitation Assessment Program, Washington, DC.

Boynton, W.R., Garber, J.H., Summers, R., Kemp, W.M., 1995. Inputs, transformations, and transport of nitrogen and phosphorus in Chesapeake Bay and selected tributaries. Estuaries 18, 285–314.

Butler, T.J., Likens, G.E., Stunder, B.J., 2001. Regional-scale impacts of Phase I of the Clean Air Act Amendments: the relationship between emissions and concentrations, both wet and dry. Atmospheric Environment 35, 1015–1028.

Castro, M.S., Driscoll, C.T., Jordan, T.E., Reay, W.G., Boynton, W.R. Nitrogen inputs to thirty-four watershed-estuary systems on the East and Gulf Coasts of the United States. Estuaries (in press).

Craig, B.W., Friedland, A.J., 1991. Spatial patterns in forest composition and standing dead red spruce in montane forests of the Adirondacks and northern Appalachians. Environmental Monitoring and Assessment 18, 129–140.

Cronan, C.S., Grigal, D.F., 1995. Use of calcium/aluminum ratios as indicators of stress in forest ecosystems. Journal of Environmental Quality 24, 209–226.

Cronan, C.S., Schofield, C.L., 1990. Relationships between aqueous aluminum and acidic deposition in forested watersheds of North America and Northern Europe. Environmental Science and Technology 24, 1100–1105.

DeHayes, D.H., Schaberg, P.G., Hawley, G.J., Strimbeck, G.R., 1999. Acid rain impacts calcium nutrition and forest health. BioScience 49, 789–800.

D'Elia, C.F., Harding Jr., L.W., Leffler, M., Mackiernan, G.B., 1992. The role and control of nutrients in Chesapeake Bay. Water Science Technology 26, 2635–2644.

Driscoll, C.T., Postek, K.M., 1995. The chemistry of aluminum in surface waters. In: Sposito, G. (Ed.), The Environmental Chemistry of Aluminum. Lewis Publishers, Chelsea, MI, pp. 363–418.

Driscoll, C.T., Blette, V., Yan, C., Schofield, C.L., Munson, R., Holsapple, J., 1995. The role of dissolved organic carbon in the chemistry and bioavailability of mercury in remote Adirondack lakes. Water Air and Soil Pollution 80, 499–508.

Driscoll, C.T., Johnson, N.M., Likens, G.E., Feller, M.C., 1988. The effects of acidic deposition on stream water chemistry: a comparison between Hubbard Brook, New Hampshire and Jamieson Creek, British Columbia. Water Resources Research 24, 195–200.

Driscoll, C.T., Lawrence, G.B., Bulger, A.J., Butler, T.J., Cronan, C.S., Eagar, C., Lambert, K.F., Likens, G.E., Stoddard, J.L., Weathers, K.C., 2001. Acidic deposition in the northeastern US: sources and inputs, ecosystems effects, and management strategies. BioScience 51, 180–198.

Driscoll, C.T., Newton, R.M., Gubala, C.P., Baker, J.P., Christensen, S., 1991. Adirondack Mountains. In: Charles, D.F. (Ed.), Acidic Deposition and Aquatic Ecosystems: Regional Case Studies. Springer-Verlag, New York, pp. 133–202.

Driscoll, C.T., Postek, K.M., Mateti, D., Sequeira, K., Aber, J.D., Kretser, W.J., Mitchell, M.J., Raynal, D.J., 1998. The response of lake water in the Adirondack Region of New York to changes in acidic deposition. Environmental Science and Policy 1, 185–198.

Driscoll, C.T., Yan, C., Schofield, C.L., Munson, R., Holsapple, J., 1994. The mercury cycle and fish in the Adirondack lakes. Environmental Science and Technology 28, 136. A-143A.

Drohan, P.J., Stout, S.L., Petersen, G.W., 1999. Spatial relationships between sugar maple (Acer saccharum Marsh.), sugar maple decline, slope, aspect, and topographic position in northern Pennsylvania. In: Horsley, S.B., Long, R.P. (Eds.), Sugar Maple Ecology and Health: Proceedings of an International Symposium (General Technical Report NE-261). US Department of Agriculture, Forest Service, Radnor, PA, pp. 46–54.

Fenn, M.E., Poth, M.A., Aber, J.D., Baron, J.S., Bormann, B.T., Johnson, D.W., Lemly, A.D., McNulty, S.G., Ryan, D.F., Stottlemyer, R., 1998. Nitrogen excess in North American Ecosystems: predisposing factors, ecosystem responses, and management strategies. Ecological Applications 8, 706–733.

Fisher, D.C., Oppenheimer, M., 1991. Atmospheric nitrogen deposition and the Chesapeake Bay estuary. Ambio 20, 102–108.

Gallagher, J., Baker, J. 1990. Current status of fish communities in Adirondack Lakes. In: Adirondack Lakes Survey: An Interpretive Analysis of Fish Communities and Water Chemistry, 1984–1987. Adirondacks Lakes Survey Corporation, Ray Brook, NY, pp. 3-11–3-48.

Gbondo-Tugbawa, S.S., Driscoll, C.T., Mitchell, M.J., Aber, J.D., Likens, G.E., 2002. A model to simulate the response of a northern hardwood forest ecosystem to changes in S deposition. Ecological Applications 12, 8–23.

Hallegraeff, G., 1993. A review of harmful algal blooms and their apparent global increase. Phycologia 32, 79–99.

Horsley, S.B., Long, R.P., Bailey, S.W., Hallet, R.A., Hall, T.J., 1999. Factors contributing to sugar maple decline along topographic gradients on the glaciated and unglaciated Alleghany Plateau. In: Horsley, S.B., Long, R.P. (Eds.), Sugar Maple Ecology and Health: Proceedings of an International Symposium (General Technical Report NE-261). US Department of Agriculture, Forest Service, Radnor, PA, pp. 60–62.

Horsley, S.B., Long, R.P., Bailey, S.W., Hall, T.J., 2000. Factors associated with the decline of sugar maple on the Allegheny Plateau. Canadian Journal of Forest Research 30, 1365–1378.

Houston, D.R., 1999. History of sugar maple decline. In: Horsley, S.B., Long, R.P. (Eds.), Sugar Maple Ecology and Health: Proceedings of an International Symposium (General Technical Report NE-261). US Department of Agriculture, Forest Service, Radnor, PA, pp. 9–26.

Ito, M., Mitchell, M.J., Driscoll, C.T., 2002. Spatial patterns of precipitation quantity and chemistry and air temperature in the Adirondack region of New York. Atmospheric Environment 36, 1051–1062.

Jaworski, N.A., Howarth, R.W., Hetling, L.J., 1997. Atmospheric deposition of nitrogen oxides onto the landscape contributes to coastal eutrophication in the Northeast United States. Environmental Science and Technology 31, 1995–2005.

Johnson, D.W., Mitchell, M.J., 1998. Responses of forest ecosystems to changing sulfur inputs. In: Maynard, D. (Ed.), Sulfur in the Environment. Marcel Dekker, New York, pp. 219–262.

Kaufmann, P.R., Herlihy, A.T., Elwood, J.W., Mitch, M.E., Overton, W.S., Sale, M.J., Messer, J.J., Cougan, K.A., Peck, D.V., Reckhow, K.H., Kinney, A.J., Christie, S.J., Brown, D.D., Hagley, C.A., Jager, H.I., 1988. Chemical Characteristics of Streams in the Mid-Atlantic and Southeastern United States. Volume 1: Population Descriptions and Physico-chemical Relationships. EPA/600/3-88/021a. US Environmental Protection Agency, Washington, DC.

Kretser, W., Gallagher, J., Nicolette, J., 1989. Adirondack Lakes Study. 1984–1987. An Evaluation of Fish Communities and Water Chemistry. Adirondacks Lakes Survey Corporation, Ray Brook, NY.

Lapointe, B.E., Matzie, W.R., 1996. Effects of stormwater nutrient discharges on eutrophication processes in nearshore waters of the Florida keys. Estuaries 19, 422–435.

Lawrence, G.B., David, M.B., Lovett, G.M., Murdoch, P.S., Burns, P.A., Baldigo, B.P., Thompson, A.W., Porter, J.H., Stoddard, J.L., 1999. Soil calcium status and the response of stream chemistry to changing acidic deposition rates in the Catskill Mountains of New York. Ecological Applications 9, 1059–1072.

Lee, V., Olsen, S., 1985. Eutrophication and management initiatives for the control of nutrient inputs to Rhode Island coastal lagoons. Estuaries 8, 191–202.

Likens, G.E., Butler, T.J., Buso, D.C., 2000. Long- and short-term changes in sulfate deposition: effects of the 1990 Clean Air Act Amendments. Biogeochemistry 52, 1–11.

Likens, G.E., Driscoll, C.T., Buso, D.C., Mitchell, M.J., Lovett, G.M., Bailey, S.W., Siccama, T.G., Reiners, W.A., Alewell, C., 2002. The biogeochemistry of sulfur at Hubbard. Biogeochemistry 60, 235–316.

Likens, G.E., Driscoll, C.T., Buso, D.C., Siccama, T.G., Johnson, C.E., Lovett, G.M., Fahey, T.J., Reiners, W.A., Ryan, D.F., Martin, C.W., Bailey, S.W., 1998. The biogeochemistry of calcium at Hubbard. Biogeochemistry 41, 89–173.

Likens, G.E., Driscoll, C.T., Buso, D.C., 1996. Long-term effects of acid rain: response and recovery of a forested ecosystem. Science 272, 244–246.

MacAvoy, S.E., Bulger, A.J., 1995. Survival of brook trout (*Salvelinus fontinalis*) embryos and fry in streams of different acid sensitivity in Shenandoah National Park, USA. Water Air and Soil Pollution 85, 445–450.

Malm, W.C., Sisler, J.F., Huffman, D., Eldred, R.A., Cahill, T.A., 1994. Spatial and seasonal trends in particle concentration and optical extinction in the United States. Journal of Geophysical Research 99, 1347–1370.

Mitchell, M.J., Driscoll, C.T., Owen, J.S., Schaefer, D., Michener, R., Raynal, D.J., 2001. Nitrogen biogeochemistry of three hardwood ecosystems in northern hardwood forest soils. Soil Science Society of American Journal 65, 93–133.

Moore, M.V., Pace, M.L., Mather, J.R., Murdoch, P.S., Howarth, R.W., Folt, C.L., Chen, C.Y., Hemond, H.F., Flebbe, P.A., Driscoll, C.T., 1997. Potential effects of climate change on freshwater ecosystems of the New England/Mid-Atlantic region. Hydrological Processes 11, 925–947.

National Acid Precipitation Assessment Program (NAPAP), 1998. NAPAP Biennial Report to Congress: An Integrated Assessment. US National Acid Precipitation Assessment Program, Silver Spring, MD.

Nixon, S., 1986. Nutrient dynamics and productivity of marine coastal waters. In: Clayton, B., Behbehani, M. (Eds.), Coastal Eutrophication. Alden Press, Oxford, pp. 97–115.

Nixon, S., 1995. Coastal marine eutrophication: a definition, social causes and future concerns. Ophelia 41, 199–220.

Paerl, H., 1988. Nuisance phytoplankton blooms in coastal, estuarine and inland waters. Limnology and Oceanography 33, 823–847.

Paerl, H., 1995. Coastal eutrophication in relation to atmospheric nitrogen deposition: current perspectives. Ophelia 41, 237–259.

Paerl, H., 1997. Coastal eutrophication and harmful algal blooms: importance of atmospheric deposition and groundwater as "new" nitrogen and other nutrient sources. Limnology and Oceanography 42, 1154–1162.

Peierls, B., Caraco, N., Pace, M., Cole, J.J., 1991. Human influence on river nitrogen. Nature 350, 386–387.

Raynal, D.J., Joslin, J.D., Thornton, F.C., Schaedle, M., Henderson, G.S., 1990. Sensitivity of tree seedlings to aluminum: III. Red spruce and loblolly pine. Journal of Environmental Quality 19, 180–187.

Ryther, J., Dunstan, W., 1971. Nitrogen, phosphorus and eutrophication in the coastal marine environment. Science 171, 1008–1112.

Schindler, D.W., Mills, K.H., Malley, D.F., Findlay, S., Shearer, J.A., Davies, I.J., Turner, M.A., Lindsey, G.A., Cruikshank, D.R., 1985. Long-term ecosystem stress: effects of years of experimental acidification. Canadian Journal of Fisheries and Aquatic Science 37, 342–354.

Shortle, W.C., Smith, K.T., 1988. Aluminum-induced calcium deficiency syndrome in declining red spruce trees. Science 240, 1017–1018.

Stoddard, J.L., 1994. Long-term changes in watershed retention of nitrogen: Its causes and aquatic consequences. In: Baker, L.A. (Ed.), Environmental Chemistry of Lakes and Reservoirs, Advances in Chemistry Series No. 237. American Chemical Society, Washington, DC.

Stoddard, J.L., Jeffries, D.S., Lukewille, A., Clair, T.A., Dillon, P.J., et al., 1999. Regional trends in aquatic recovery from acidification in North America and Europe. Nature 401, 575–578.

Valiela, I., Costa, J.E., 1988. Eutrophication of Buttermilk Bay, a Cape Cod coastal embayment: concentrations of nutrients and watershed nutrient budgets. Environmental Management 12, 539–553.

Valiela, I., Costa, J.E., Foreman, K., Teal, J.M., Howes, B., Aubrey, D., 1990. Transport of groundwater-borne nutrients from watersheds and their effects on coastal waters. Biogeochemistry 10, 177–197.

Van Sickle, J., Baker, J.P., Simonin, H.A., Baldigo, B.P., Kretser, W.A., Sharpe, W.F., 1996. Episodic acidification of small streams in the northeastern United States: fish mortality in field bioassays. Ecological Applications 6, 408–421.

Vitousek, P.M., Aber, J.D., Howarth, R.W., Likens, G.E., Matson, P.A., Schindler, D.W., Schlesinger, W.H., Tilman, G.D., 1997. Human alterations of the global nitrogen cycle: causes and consequence. Issues in Ecology 1, 1–15.

Wigington Jr., P.J., Baker, J.P., DeWalle, D.R., Kretser, W.A., Murdoch, P.S., Simonin, H.A., Van Sickle, J., McDowell, M.K., Peck, D.V., Barchet, W.R., 1996. Episodic acidification of small streams in the northeastern United States: Episodic Response Project. Ecological Applications 6, 374–388.