Effects of Air Pollution on Ecosystems and Biological Diversity in the Eastern United States

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Conservation organizations have most often focused on land-use change, climate change, and invasive species as prime threats to biodiversity conservation. Although air pollution is an acknowledged widespread problem, it is rarely considered in conservation planning or management. In this synthesis, the state of scientific knowledge on the effects of air pollution on plants and animals in the Northeastern and Mid-Atlantic regions of the United States is summarized. Four air pollutants (sulfur, nitrogen, ozone, and mercury) and eight ecosystem types ranging from estuaries to alpine tundra are considered. Effects of air pollution were identified, with varying levels of certainty, in all the ecosystem types examined. None of these ecosystem types is free of the impacts of air pollution, and most are affected by multiple pollutants. In aquatic ecosystems, effects of acidity, nitrogen, and mercury on organisms and biogeochemical processes are well documented. Air pollution causes or contributes to acidification of lakes, eutrophication of estuaries and coastal waters, and mercury bioaccumulation in aquatic food webs. In terrestrial ecosystems, the effects of air pollution on biogeochemical cycling are also very well documented, but the effects on most organisms and the interaction of air pollution with other stressors are less well understood. Nevertheless, there is strong evidence for effects of nitrogen deposition on plants in grasslands, alpine areas, and bogs, and for nitrogen effects on forest mycorrhizae. Soil acidification is widespread in forest ecosystems across the eastern United States and is likely to affect the composition and function of forests in acid-sensitive areas over the long term. Ozone is known to cause reductions in photosynthesis in many terrestrial plant species. For the most part, the effects of these pollutants are chronic, not acute, at the exposure levels common in the eastern United States. Mortality is often observed only at experimentally elevated exposure levels or in combination with other stresses such as drought, freezing, or pathogens. The notable exceptions are the acid/aluminum effects on aquatic organisms, which can be lethal at levels of acidity observed in many surface waters in the region. Although the effects are often subtle, they are important to biological conservation. Changes in species composition caused by terrestrial or aquatic acidification or eutrophication can propagate throughout the food webs to affect many organisms beyond those that are directly sensitive to the pollution. Likewise, sublethal

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doses of toxic pollutants may reduce the reproductive success of the affected organisms or make them more susceptible to potentially lethal pathogens. Many serious gaps in knowledge that warrant further research were identified. Among those gaps are the effects of acidification, ozone, and mercury on alpine systems, effects of nitrogen on species composition of forests, effects of mercury in terrestrial food webs, interactive effects of multiple pollutants, and interactions among air pollution and other environmental changes such as climate change and invasive species. These gaps in knowledge, coupled with the strong likelihood of impacts on ecosystems that have not been studied in the region, suggests that current knowledge underestimates the actual impact of air pollutants on biodiversity. Nonetheless, because known or likely impacts of air pollution on the biodiversity and function of natural ecosystems are widespread in the Northeast and Mid-Atlantic regions, the effects of air pollution should be considered in any long-term conservation strategy. It is recommended that ecologically relevant standards, such as "critical loads," be adopted for air pollutants and the importance of long-term monitoring of air pollution and its effects is emphasized.

Key words: **acid deposition; sulfur; nitrogen; mercury; ozone; eastern United States; forest; stream; lake**

Introduction

Background

The past four decades have seen much research on the effects of air pollution on ecosystems. Much of this work has focused on biogeochemical responses of lakes, forests, and estuaries to elevated levels of sulfur and nitrogen deposition. Many studies have shown that plant and animal species differ in their sensitivity to air pollution and its biogeochemical consequences, such as soil and water acidification. For instance, lichens differ in sensitivity to sulfur dioxide (e.g., Thormann 2006), vascular plants differ in sensitivity to ozone (e.g., King *et al.* 2005), fish and other aquatic biota differ in sensitivity to lake acidity (e.g., Baker *et al.* 1990), and grasses and forbs differ in sensitivity to added nitrogen (e.g., Wedin & Tillman 1996). This differential sensitivity suggests that air-pollution stress will have consequences for the biodiversity of an ecosystem, either in shifting species composition or outright loss of sensitive species. Because these impacts are species-specific and pollutant-specific, and most ecosystems are exposed to multiple air pollutants simultaneously, it can be difficult to assess the overall impact of air pollution on organisms in a particular ecosystem. This lack of a "big picture" makes it difficult for conservation biologists and policymakers to determine the role of air pollution among the array of threats to biodiversity.

This report is a synthesis of scientific information on the impacts of air pollution on elements of biological diversity (e.g., species, natural communities, ecosystems) in the Northeastern and Mid-Atlantic regions of the United States. These elements are used by The Nature Conservancy (TNC), the largest conservation organization in the United States, to best represent biological diversity in planning efforts used to guide and focus conservation action. By focusing on TNC targets, we hope to provide conservation organizations with sufficient information to gauge the scope and severity of the threat of air pollution in this region. Our approach in this synthesis is to group species and communities in eight ecosystem types and to assess the evidence for the effects of four key air pollutants in each ecosystem type. To the extent possible, we attempt to quantify the levels of air pollution that produce adverse impacts, and to assess the certainty of scientific knowledge on the subject. Our goal is to gain a better appreciation for the combined impacts of air pollution on multiple taxonomic groups and ecosystem types in order to better inform biodiversity-conservation efforts about the relative significance of this threat. To our knowledge, this type of collective synthesis of impacts to biological diversity has not been previously attempted for multiple pollutants across multiple conservation target groups.

Air Pollutants Considered

In this review we consider four air pollutants of concern: sulfur (S), nitrogen (N), mercury (Hg), and ozone (O_3).

Sulfur and nitrogen are primarily released from fossil-fuel combustion as S and N oxides, and these gases can be transformed in the atmosphere to acidic particles and acid precipitation (Driscoll *et al.* 2001). The gases and particles may be deposited directly to vegetation and soil surfaces in a process known as dry deposition, or they may be incorporated into cloud droplets, raindrops, or snowflakes to increase the acidity of precipitation (Lovett 1994). In mountaintop and coastal areas where cloud and fog are common, highly acidic cloud droplets can deposit directly to vegetation. Sulfur oxides are released primarily from coal combustion, whereas any combustion can produce N oxides. Thus the contribution of motor vehicles to the pollution problem is greater for N oxides than for S oxides (Weathers *et al.* 2006a).

Nitrogen can also be emitted from various agricultural activities as ammonia, a gas that can react with acidic gases and particles in the atmosphere to form small particles containing ammonium salts. Ammonia gas and ammonium particles can be dry-deposited to vegetation or can be dissolved in precipitation. In areas of intense agricultural activity (e.g., downwind of feedlots or heavily fertilized crops), ammonium can be the dominant form of N deposition (Driscoll *et al.* 2003).

The effects of S and N pollution on ecosystems are generally not caused by direct physiological effects of exposure to the gases, except in sites that are very close to emission sources. More commonly, the effects are related to the chronic accumulation of S and N in plants and soils and the long-term changes in soil and water chemistry caused by deposition of sulfate, nitrate, and ammonium. Nitrogen and S can be transported long distances in the atmosphere and can impact ecosystems hundreds of kilometers from the emission sources. Because sulfate and nitrate ions can be readily leached from soils to surface waters, deposition of these pollutants to terrestrial ecosystems may cause a cascade of effects that includes lakes, streams, rivers, estuaries, and the coastal ocean.

Ozone is a "secondary" pollutant, formed in the atmosphere from photochemical reactions involving N oxides and hydrocarbons. Ground-level O_3 is a widespread regional pollutant in the eastern United States but tends to occur in particularly high concentrations downwind of major urban areas (U.S. EPA 2006). Because O_3 and its precursors can be transported long distances, it is a threat to ecosystems far removed from urban centers. Aside from its well-known effects on the human respiratory system, O_3 is known to reduce photosynthesis in most plants and cause foliar lesions in sensitive plants (U.S. EPA 2006; see Appendix 1 in this source for a list of sensitive plant species). Its effects on animals other than humans and animals that serve as medical models for humans (e.g., Norway rats) have not been well studied but are likely to be significant (Menzel 1984).

Mercury released primarily from coal combustion, waste incineration and industrial processes, and is deposited to the Earth in precipitation as well as in gaseous and particulate dry deposition. The different chemical forms of Hg in the atmosphere have varying residence times (hours to months) and transport distances (local to global; Driscoll *et al.* 2007). Mercury is a known neurotoxin that biomagnifies in food chains and bioaccumulates in individuals, thus organisms at the highest trophic levels that live the longest have the greatest risk of high exposure (Evers *et al.* 2005). Exposure to mercury largely occurs as methylmercury, which is formed primarily by sulfate-reducing bacteria, particularly under anaerobic conditions. Such conditions are common in freshwater aquatic sediments, wetlands, or saturated soils (Wiener *et al.* 2003).

Structure of This Synthesis

We organized this chapter in a series of sections. In the second section we discuss ecosystem types and the specific biological taxa that are most closely associated with these ecosystem types. In the discussion of each taxon or ecosystem type, we attempt to synthesize: (1) the certainty or uncertainty of the scientific information on air-pollution impacts on that target; (2) the nature of those impacts; and (3) if possible, the levels of loading of the pollutant that are known to produce these impacts.

Certainty of Information

In some cases, there is much information on air-pollution effects on specific targets in the Northeast and Mid-Atlantic regions, and in other cases less information is available. In general, we reserved the highest level of confidence for targets in which air pollution impacts are well established from experimental and/or gradient studies within the region. (NOTE: Experimental studies manipulate the exposure to the pollutant in field or laboratory situations; gradient studies assess the impacts along a gradient of ambient exposure to the pollutant.) We had moderate confidence in conclusions inferred from studies on similar species or ecosystems outside the region. We had low confidence in conclusions drawn from mechanistic arguments about what species are likely to be sensitive to pollution, when those arguments were accompanied by little or no direct empirical support from either inside or outside the region. We tried to assess confidence levels no matter what the level of impact (e.g., in some cases we had high confidence that there was little or no impact on a taxon or ecosystem type).

Nature of the Impacts

We considered several different types of impacts, including direct effects of pollutants on biological functioning of organisms (e.g., toxicity, mortality, effects on growth, or reproduction), effects on species composition in communities, effects on abiotic ecosystem characteristics that are likely to affect the biota over the long term, and indirect effects in which species are affected through food web or competitive interactions (e.g., negative impacts on a species may benefit its prey and its competitors). We also attempted to capture specific examples of known effects on taxa or ecosystem types to help illustrate the impacts.

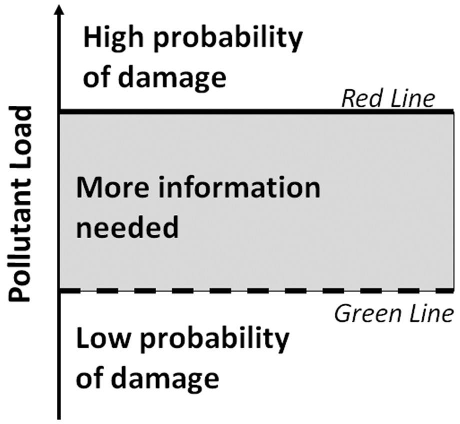

Figure 1. Red line/green line model used by Fox *et al.* (1989) to screen potential air pollution impacts on USDA Forest Service wilderness areas.

Loading Levels that Produce Impacts

In some cases we felt there was sufficient information to quantify the relationship between exposure to the pollutant and impacts on ecosystems. To do this, we followed the approach of Fox *et al.* (1989) in a report designed to help managers of U.S. Department of Agriculture (USDA) Forest Service wilderness areas determine the potential for impacts for new air pollution sources proposed for the airshed of the wilderness area. Fox *et al.* (1989) proposed determining a green line and a red line of air-pollution exposure (Fig. 1)—the green line is the deposition or concentration level below which there is high certainty that no adverse impacts will occur, and the red line is the deposition or concentration level above which there is high certainty of adverse impacts on at least some component of the ecosystem. Between the red line and the green line is the "yellow zone" where more information is needed to determine if air pollution will have a significant impact on the system—for instance, that information could be particularly characteristic of the site or the specifics of the exposure conditions. The Fox *et al.* (1989) approach recognizes the possibility of substantial variation in pollution sensitivity among ecosystems of the same type. As discussed later in the chapter, in some cases we were successful in specifying the red- and green-line values for target taxa and ecosystems, but in many cases we were not.

The ecosystem types considered in Section 2 are as follow.

Terrestrial Ecosystems (including embedded freshwater wetlands):

- alpine and subalpine systems,
- forests (both upland and wetland types),
- bogs and fens,
- grasslands.

Aquatic Ecosystems (including freshwater, brackish, and marine):

- high gradient headwater streams,
- lakes and ponds,
- low-gradient rivers,
- estuaries, bays, and salt marshes.

For each ecosystem type, we examined the effects of the four pollutants considered: sulfur, nitrogen, mercury, and ozone.

In the third section of the chapter we discuss issues that cut across the various ecosystems and are important for understanding the full impact of air pollution on biodiversity. For instance, the issues of intersystem transport of pollution and interaction among pollutants are discussed in this section. Finally, in the fourth section, we summarize the overall conclusions of the review and discuss its relevance to conservation policy.

Effects of Air Pollution

Terrestrial Ecosystems

While the public is generally aware of the effects of air pollution on aquatic ecosystems because of media coverage of acid rain–damaged lakes and Hg-contaminated fish, the public is much less aware of effects on terrestrial ecosystems. In general, the biogeochemical impacts of S and N on ecosystems depend upon the mobility of these pollutants in the canopy and soils to which they are deposited. If the anions (negatively charged ions) they form (sulfate and nitrate) are leached through the canopy and soils, rather than being retained, they can strip the foliage and soils of valuable nutrient cations (positively charged ions) such as calcium and magnesium (Fig. 2). The leaching of sulfate and nitrate results in acidification of soils and surface waters, and in some cases mobilizes aluminum. Aluminum is a natural component of soils, but under acid conditions it becomes more soluble and exhibits high concentrations in soil water, where it can be toxic to roots, and it can leach into surface waters where it is toxic to fish and other aquatic organisms.

Both S and N can also accumulate in the vegetation and soil, leading to delayed effects as the accumulated S or N is slowly released to water for years or decades after its initial deposition. Accumulation of N in terrestrial ecosystems can cause shifts in species composition as N-loving species outcompete those species better adapted to less fertile soils (Gough *et al.* 2000). Nitrogen accumulation may also lead to a condition known as N saturation, in which overabundance of this key nutrient results in a series of impacts on microbial and plant production and N cycling (e.g., Aber *et al.* 1998).

Ozone is a well-studied pollutant known to be toxic to plants and animals. In plants, O_3 appears to affect membrane function, leading to reduction in photosynthesis, slower growth, and in severe cases, death. In animals, O_3 effects have mainly been studied in humans, where it damages lung tissue and exacerbates

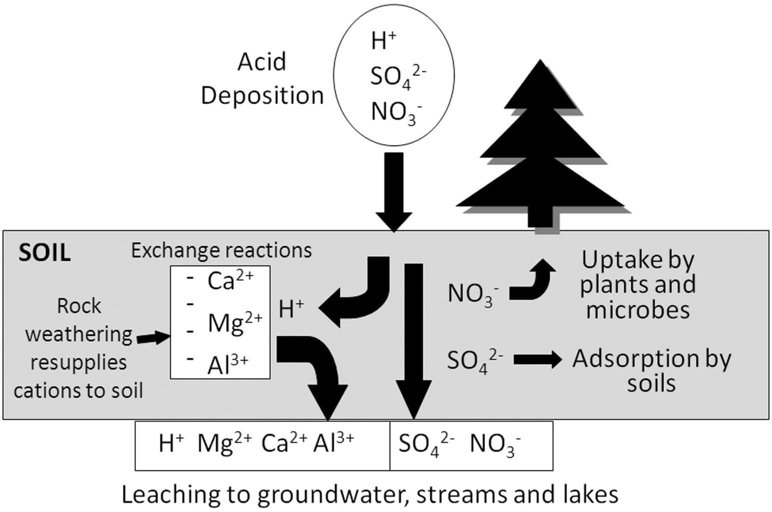

Figure 2. Schematic of key interactions of acidic deposition with soils. Biological (uptake), soil (exchange and adsorption) and geological (weathering) factors interact to determine the effects of the acidic deposition in the soil.

respiratory problems such as asthma (U.S. EPA 2006).

Mercury is known to accumulate in soils, but studies of its effects have primarily focused on aquatic ecosystems where anaerobic conditions facilitate the production of a form called methylmercury (Driscoll *et al.* 2007). Exposure of humans and animals is largely associated with methylmercury. Previously, the terrestrial organisms considered to be at risk for mercury contamination were animals that feed on other animals from the aquatic food web, such as birds feeding on aquatic insects or raccoons that eat aquatic invertebrates. Only recently have researchers begun to examine the methylation and bioaccumulation of Hg in terrestrial food webs, and more conclusive information is likely forthcoming in the next few years.

In the following sections, we highlight specific ecosystem types, summarizing what is known about air pollution effects in those ecosystems, and also identifying many areas where additional research is warranted.

Alpine Ecosystems

Nitrogen. We know of no direct studies from the eastern United States on the effects of N deposition on the herbaceous and shrub communities that constitute the alpine zone of northeastern mountains. However, there have been studies on the effects of N on alpine ecosystems in the Rocky Mountains and in Europe. At Niwot Ridge in the Rocky Mountains of Colorado, an N enrichment study in an alpine meadow showed that N addition increases overall plant diversity, primarily by increasing the abundance of a sedge (*Carex rupestris*), and several other species (Bowman *et al.* 2006). This study concluded that species composition is a more sensitive indicator of changes due to N deposition than is soil chemical response, and the plant responses are evident at N deposition rates as low as 4 kg N/ha-y. The authors speculated that higher levels of N deposition or long-term accumulation of N in the ecosystem may cause a decrease in plant-species diversity as nitrophilic species start to dominate over those species less responsive to N (Bowman *et al.* 2006).

Because of the effects of N shown in the experiments in the Rockies and the overall floristic and structural similarity of alpine ecosystems in the Rockies and the eastern United States, we have moderate confidence that N deposition is affecting alpine ecosystems in the eastern United States. Deposition loads in eastern alpine zones probably range from 10 to 20 kg N/ha-y (Ollinger *et al*. 1993; Weathers *et al*. 2000), and have probably been at that level for several decades, so it is possible that productivity and species shifts have already occurred in these ecosystems. However, in the absence of direct experimental evidence, gradient studies, or long-term monitoring of vegetation in these ecosystems, the nature and magnitude of the effects remain highly uncertain.

Sulfur, Acidity, Mercury, and Ozone. We know of no direct studies of the effects of S deposition, acid deposition, Hg, or O_3 on alpine ecosystems by this group, either in the eastern or western United States. Because of their high elevation, these ecosystems are exposed to high deposition rates of all of these pollutants (Lovett and Kinsman 1990; Aneja *et al.* 1994; Weathers *et al.* 2006b). For red spruce trees in isolated patches within the alpine zone, one might expect sensitivity to acid-induced calcium leaching as for red spruce in forests (see discussion in the Forest section); however, this has not been demonstrated experimentally in alpine red spruce. Most alpine plants have never been tested for sensitivity to O_3. Likewise, we know of no studies of Hg accumulation in food chains in alpine ecosystems. Small wetlands and frequently saturated soils in alpine ecosystems may offer an opportunity for methylation that transforms ionic Hg into highly bioavailable methylmercury. Thus, for all of these pollutants we consider the potential exposure levels to be high and impacts to be likely, but we have low confidence in any prediction of specific impacts because of the lack of relevant studies.

Bogs and Fens

Nitrogen. Bogs and fens may be among the most sensitive ecosystems to the eutrophication effects of N deposition because they tend to be nutrient-poor, and ombrotrophic bogs in particular receive all of their nutrients from atmospheric deposition. Bogs and fens are listed by Bobbink *et al.* (1998) as being among the ecosystems at highest risk of species compositional shifts due to N deposition. This subject has received extensive research attention in Europe, where increases in N deposition have been associated with decline in typical bog species such as the sundew (*Drosera)* and certain species of *Sphagnum*. Often favored are graminoids such as *Deschampsia* and *Eriophorum* that can grow tall and outcompete the bog species for light. Bedford *et al.* (1999) suggest that most North American wetlands are more likely to be limited by phosphorus (P) than by N, but marshes and swamps are the wetland types most likely to show N limitation or N and P colimitation. However, species adapted to low-N environments may be quite sensitive to inputs of added N. An N enrichment study in bogs in New England showed substantial effects on growth and reproduction of the pitcher plant (*Sarracenia purpurea*) (Ellison and Gotelli 2002). If there is no change in current N deposition rates, these changes suggest a substantial probability of extinction of local populations within 100–250 years (Gotelli and Ellison 2002, 2006).

Recent results reported from a long-term Canadian experiment suggest that chronic N deposition may initially lead to enhanced carbon uptake as the microbial community changes and decomposition slows (Basiliko *et al.* 2006). However, in the longer term, there may be a shift to decreased carbon sequestration and increases in CO_2 and CH_4 emissions as vegetation changes associated with higher N availability lead to production of plants that decompose more rapidly. These results, combined with the accumulated evidence from Europe and elsewhere and the direct experimental evidence from New England, give us high confidence that N deposition will strongly affect bog and fen ecosystems. Given the sensitivity of bog ecosystems to N enrichment, it is likely that N effects are already occurring in many areas of the eastern United States.

Sulfur and Acidity. Several biogeochemical responses of wetlands to S deposition are important and well understood. As more sulfate is deposited, the activity of sulfate-reducing bacteria is stimulated. These bacteria gain energy from the chemical reduction of sulfate to sulfide in anaerobic conditions such as those that occur in wetland soils and sediments. Because these bacteria also methylate Hg, the increase in their activity increases the potential for the formation of methylmercury. This interaction between the S and Hg cycles results in more formation of bioavailable methylmercury in response to increases in S deposition (Jeremiason *et al.* 2006; Branfireun *et al.* 1999). The increase in sulfate-reducing bacteria also consumes labile carbon and hydrogen, reducing their availability to methanogenic bacteria and thus decreasing methane production by wetlands (Dise and Verry 2001).

Mercury. We are not aware of any direct study of Hg on the organisms of bogs and fens. However, the role of wetlands in Hg cycling is well known (Wiener *et al.* 2003). Because of the anaerobic conditions in their soils and sediments, wetlands are hot spots for methylation of Hg in the landscape (Grigal 2003; Evers *et al.* 2007). Thus the ionic Hg draining from a watershed can be converted to methylmercury in a wetland, where it can then be transported to a stream or lake or consumed in the wetland and transferred up the food chain. We would expect the higher trophic–level consumers in bogs and fens (e.g., birds that consume bog insects) to be at high risk for Hg accumulation.

Ozone. The few studies of the effects of O_3 on the plants of bog and fen ecosystems show that wetland plant species vary considerably in their sensitivity to O_3. Ozone exposure studies in Finland show the important sedge *Eriophorum vaginatum* to be relatively insensitive to ozone; however, several of the tree species that are commonly found in or around wetlands in the eastern United States are considered sensitive to O_3 [e.g., green ash (*Fraxinus pennsylvanica*), speckled alder (*Alnus rugosa*)] (National Park Service 2003). Even within the important *Sphagnum* genus of bryophytes, some species are sensitive to O_3 and some are not (Gagnon and Karnosky 1992; Potter *et al.* 1996.) This range in response suggests that some wetlands ecosystems may be negatively affected by O_3, and further that O_3 may shift plant community composition in favor of more tolerant species.

Forests

Forests have received more air pollution–related research attention than any other terrestrial ecosystem type. However, the studies have largely focused on biogeochemical responses, and the links to species composition are often unclear. This is probably because the dominant organisms in forests—trees—are so long-lived that studying the population and community responses to a chronic stress such as air pollution requires long-term research. Nonetheless, some recent studies have begun to elucidate actual and potential biological responses.

Nitrogen. There has been much research on the effects of N deposition on forests, both within the eastern United States and elsewhere, particularly the western United States and western Europe. This research has caused a major shift in the perceptions of N by forest ecologists. Previously, N was considered solely as a limiting nutrient for forest production, and fertilization with N was used to enhance production. In the last 20 years, however, research has shown that chronic N addition can have toxic effects that alter plant, soil, and microbial interactions, and can lead to loss of soil fertility, reduced productivity, and even tree death. The basic processes involved have been organized in a conceptual framework referred to as “nitrogen saturation” (e.g., Aber *et al.* 1998). While this is still an active area of research, most scientists agree that continued accumulation of N in terrestrial ecosystems causes significant responses in ecosystem function.

Temperate-zone forests in unpolluted areas are usually limited by N, which means that additions of N can stimulate productivity. Most of the N that is deposited from the atmosphere is taken up by plants or microbes and retained in the vegetation or in soil organic matter. Leaching of N from these forests in drainage water is usually minimal, except in the case of older

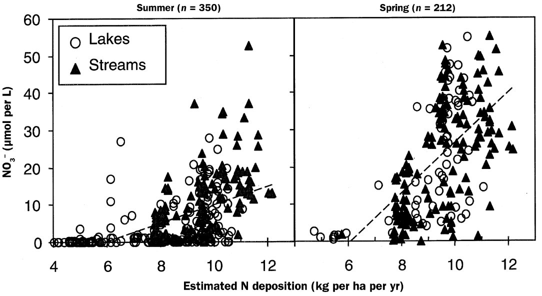

Figure 3. Nitrate concentration in surface waters of the northeastern U.S. in spring (left panel) and summer (right panel) vs. estimated N deposition. In the legend, WV = West Virginia, CAT = Catskill Mts., ADK = Adirondack Mts., VT = Vermont, NH = New Hampshire, and ME = Maine. From Aber *et al.* (2003). Reproduced with the permission of **Bioscience**.

or damaged forests, which have reduced N uptake capacity because of slower growth rates. The conceptual model of nitrogen saturation (Aber *et al.* 1998) predicts that as deposition of N increases due to air pollution, N accumulates in the soils and vegetation. The accumulation of N increases the opportunity for N leaching, which occurs primarily as the mobile anion nitrate. Foliar and wood N concentrations increase, and the microbial processes that transform N in the soil are enhanced. In the United States, there is little evidence for increased tree growth from the fertilizing effect of this added N, probably because most of the N appears to be retained in the soil organic matter where it is unavailable to the plants (Nadelhoffer *et al.* 1999; Templer *et al.* 2005). However, increased nitrate leaching is observed in some (but not all) forested ecosystems in the Northeast as N deposition levels increase (Aber *et al.* 2003) (Fig. 3). The differences between those ecosystems that show increased N leaching and those that do not is probably related to forest disturbance history (e.g., fire, logging, agriculture.), tree species composition, and soil properties, especially the soil carbon:nitrogen ratio (Goodale and Aber 2001; Lovett *et al.* 2002). Similar results have been found in Europe, where the most important factors controlling the amount of nitrate leaching from forests are the amount of N deposition and the carbon:nitrogen ratio of the forest floor (MacDonald *et al.* 2002). Nitrate leaching is important because it acidifies soils, depleting important nutrients such as calcium and magnesium and mobilizing aluminum. In this regard, nitrate acts in concert with sulfate leaching, thus N and S pollution can have additive effects.

The effects of N deposition on plant-species composition of forests are not as well studied as the biogeochemical effects. Shifts in tree species composition under ambient deposition levels would be difficult to assess because of the long generation time of trees. Even if long-term studies revealed such shifts, it would be difficult to attribute these changes unambiguously to N deposition to eastern forests, which are simultaneously being exposed to many new stresses (e.g., O_3, climate change, exotic pests) in addition to N deposition. Changes in abundance and composition of understory shrubs and herbs might also be expected in response to N deposition, as has been shown in Europe (Bobbink *et al.* 1998). In the eastern United States the data are not as clear;the few studies that have examined this effect have used fertilization experiments as opposed to gradient studies or long-term measurements, and the studies have shown varying responses (Jordan *et al.* 1997; Hurd *et al.* 1998; Rainey *et al.* 1999; Gilliam *et al.* 2006). It is known from studies in the eastern United States that N addition shifts the activity of soil microorganisms, with some responding positively to N addition, while others respond negatively (e.g., Carreiro *et al.* 2000). Across a gradient of N deposition in the eastern United States, changes in microbial N cycling activity are seen in some forest types but not others (McNulty *et al.* 1991; Lovett and Rueth 1999). Shifts in abundance and species composition of mycorrhizal fungi, which form the crucial interface between roots and soil for most plants, have been observed in response to N deposition in Europe, Alaska, and southern California (Arnolds 1991; Lilleskov *et al.* 2001; Siguenza *et al.* 2006). Herbivorous insects tend to prefer plants with higher N concentration, and there is some evidence that increased N may be predisposing trees to attack by insect pests such as the hemlock woolly adelgid (*Adelges tsugae*) (e.g., McClure 1991) and the beech scale (*Cryptococcus fagisuga*) (Latty *et al.* 2003). Increased susceptibility to pests could be a serious liability for eastern forests, given the number of exotic insect pests that are being introduced continually through enhanced global trade (Lovett *et al.* 2006).

In the last 15 years, experimental studies of N addition to forest stands or watersheds have been reported from Maine, New Hampshire, Vermont, Massachusetts, New York, and West Virginia (Norton *et al.* 1994; Mitchell *et al.* 1994; McNulty *et al.* 1996; Adams *et al.* 1997; Magill *et al.* 1997; Templer *et al.* 2005; Adams *et al.* 2006). The N application rates vary from about 2–15 times ambient N deposition levels. A few stands show nearly complete retention of N with little biological or biogeochemical response, but most show increases in plant N content, microbial N cycling, production and leaching of nitrate, and leaching of cations such as magnesium and calcium. In three cases, a high-elevation spruce–fir forest on Mt. Ascutney, Vermont, a red pine forest in central Massachusetts, and a mixed-oak forest in southern New York, the N addition resulted in declines in productivity and increases in tree mortality (McNulty *et al.* 1996; Magill *et al.* 1997; Wallace *et al.* 2007). The mechanism of this effect is not yet understood, but in all three cases soil acidification and the resulting aluminum toxicity to roots is a strong possibility (Aber *et al.* 1998; Wallace *et al.* 2007). It is alarming to see tree mortality in response to the addition of a nutrient that was previously thought to be beneficial, but it must be remembered that these are experiments with artificially enhanced N deposition, and mortality does not appear to be a widespread response to N deposition under the current ambient deposition loads in the eastern United States. What remains unclear, and will be extremely important to resolve, is whether forests will respond the same way to long-term accumulation of N from atmospheric deposition as observed in relatively short-term experimental N additions.

Little is known about the response of forest animals in the eastern United States to N deposition, although one might expect that increases in the N content of plant tissue and shifts in soil microbial activity would cause subtle ramifications throughout the aboveground and belowground food webs.

We have high confidence in the conclusion that N deposition produces both biogeochemical and biological effects in forests. The best available criteria for setting red- and green-line values is nitrate leaching, which appears to be very rare in forests receiving <5 kg N/ha-y and increasingly common as deposition levels increase beyond 8 kg/ha-y (Fig. 3). Similar thresholds have been reported for European forests (e.g., Dise and Wright 1995). Because we considered the nitrate leaching as a symptom that indicates current microbial response and may result in vascular plant response over a time frame of decades, we estimated the green- and red-line values at 5 and 8 kg N/ha-y, respectively.

Sulfur. Much research has also been done with regard to the effects of S in forest ecosystems. Because S is usually not a biologically limiting element, the responses are in many ways less complex than those of N. Atmospherically deposited sulfate enters plant and microbial pools, but the S itself does not appear to cause any direct biological responses other than the stimulation of S-reducing bacteria in anaerobic environments (discussed earlier). Because S deposition in the eastern United States far exceeds the biological requirement for the element, most of the deposited S is either leached from the ecosystem or retained in the soils in both inorganic and organic forms (Likens *et al.* 2002). In glaciated regions of the Northeast, soils have little sulfate adsorption capacity, so most of the deposited sulfate leaches through the canopy and soils, stripping nutrient cations such as calcium and magnesium in the process. The resulting acidification of the soils mobilizes aluminum, which can be toxic to tree roots and, when it enters surface waters, to fish and other aquatic organisms (Cronan and Grigal 1995; Driscoll *et al.* 2001).

In unglaciated areas, the sulfate leaching is mitigated to varying extent by sulfate adsorption in the subsoil, although even the low levels of sulfate leaching in these ecosystems can acidify streams in sensitive areas (Galloway *et al.* 1983; Webb *et al.* 1994). Some of the sulfate retained in the soil may be remobilized in the future as S deposition levels decline, leading to a long-term legacy of elevated sulfate in stream water that would slow the rate of recovery of streams in response to declining S emissions (Sullivan *et al.* 2008).

Thus the biological effects of S deposition in forests are largely due to the acidification and cation losses caused by sulfate leaching. In areas of high-base cation supply, such as areas with calcium-rich bedrock (e.g., limestone), the soil cation losses are less of a problem, and the soils and streams are well buffered against acidification. In more sensitive, base-poor areas, loss of soil cations can be a problem for plants that require a high calcium or magnesium supply, such as sugar maple (*Acer saccharum*), white ash (*Fraxinus americana*), basswood (*Tilia americana*), and flowering dogwood (*Cornus florida*). Declines in sugar maple have been observed in calcium-poor areas in central and western Pennsylvania, attributed to a combination of acid deposition and insect outbreaks (Horsley *et al.* 2002), which may be linked to increases in oxidative stress (St. Clair *et al.* 2005). Fertilization of plots with calcium and magnesium appears to reverse the decline (Long *et al.* 1997).

In ecosystems without substantial amounts of base cations in the bedrock or glacial till, replenishment of these nutrient cations by rock weathering and atmospheric deposition is a very slow process. Therefore, soil acidification and base cation depletion are long-term processes that may take decades or even centuries to reverse after the leaching losses are stopped (Driscoll *et al.* 2001). The recovery time is further lengthened by the continued slow release of accumulated S and N from within the ecosystem after emissions and deposition of S and N are controlled (Driscoll *et al.* 2001).

In the mountains of the eastern United States, red spruce has been shown to be sensitive to acid deposition, because the acidity leaches calcium from the foliage. The loss of foliar calcium, especially the small portion of that calcium that is membrane-bound, renders the tree less able to develop frost hardiness in the autumn, leading to winter damage and in many cases, tree death (DeHayes *et al.* 1999). This mechanism is thought to be responsible for the widespread spruce decline observed in northeastern mountains during the 1980s, a phenomenon that continues to this day (Hawley *et al.* 2006).

Animals may also be affected by soil acidification. Earthworms, slugs, millipedes, centipedes, collembolans, and isopods are among the soil animals known to be sensitive to acidity, although most of the studies are from high-deposition areas in Europe (Rusek and Marshall 2000). One recent study reports that productivity of wood thrush *(Hylocichla mustelina)* populations is negatively correlated with acid deposition levels across the northeastern United States (Hames *et al.* 2002). This is purely a correlative result, but the proposed mechanism for the response is plausible—that acid deposition reduces the quantity or quality of the soil invertebrates that are the main source of calcium for wood thrushes. The reduction in acid-sensitive invertebrates is also reported to have caused declines in European birds (Graveland *et al.* 1994). In an acidified forest in Pennsylvania, adding lime to increase calcium levels improved the bird habitat (Pabian and Brittingham 2007).

The levels of S deposition that result in ecological damage are determined by edaphic factors such as the soil type and bedrock mineralogy. Several types of static and dynamic models have been applied to forests throughout the region to project soil and stream acidification responses to S deposition, taking into account such crucial variables as soil and bedrock chemistry (Miller 2006; Gbondo-Tugbawa and Driscoll 2003; Cosby *et al.* 1985).

Mercury. Mercury is a widespread pollutant in the forests of the eastern United States. It can accumulate in soils and is converted to its most bioavailable form, methylmercury, in anaerobic environments such as wetlands, riparian zones, and any other upland area with moist soils (Driscoll *et al.* 2007). Plants appear to be insensitive to methylmercury, but in animals it is a potent neurotoxin that can cause physiological, behavioral, and ultimately reproductive impacts. Mercury is biomagnified in food webs; thus, animals that are at highest risk of Hg toxicity are those feeding relatively high on the food chain, especially if their food originates from habitats with moist soils or water that are conducive to methylation. Animals that are long-lived are also at high risk, particularly in individuals where the input of methylmercury exceeds its ability to depurate or demethylate Hg.

Wildlife in both wetland and upland forests were previously considered safe from the impacts of methylmercury because of conventional thought that only aquatic ecosystems have the ability to biomagnify methylmercury and that a fish-based food web was the only one of concern. Recent findings now show that species that are not linked to the fish food web can contain surprisingly elevated levels of methylmercury. Elevated methylmercury levels have been found in birds of subalpine ecosystems, such as the blackpoll warbler and the endemic Bicknell's thrush (Rimmer *et al.* 2005). Apparently, high-elevation forests not only experience high levels of Hg deposition (Miller *et al.* 2005), but also provide an environment where Hg methylation can occur. Rimmer *et al.* (2005) documented the strong and predictive relationship of litterfall Hg values modeled by Miller *et al.* (2005) and the blood Hg values of the Bicknell's thrush. A recent study demonstrates that mercury from rivers can be transferred to the adjacent terrestrial food web via predatory spiders (Cristol *et al.* 2008).

We have high confidence that Hg pollution is affecting biogeochemical processes (methylation) and animals of eastern forests. However, we did not specify red- and green-line values, because the science is still developing and is not yet quantified at the confidence level needed.

Ozone. Ozone has been the subject of much research because it is one of the federal "criteria" pollutant, that is, the concentration is regulated by the EPA, and states are required to comply with those regulations. The regulatory standards are based primarily on the effects of O_3 on human health, but effects on plants are also well known. Ozone is a potent oxidant, and once it enters a plant through stomata, it reduces photosynthesis and alters carbon allocation (U.S. EPA 2006). Ozone at the levels found in the eastern United States often does not kill plants outright but slows their growth and may make them more susceptible to other fatal stresses such as insect or pathogen attack (U.S. EPA 2006). Ozone exposure can also reduce flowering (Bergweiler and Manning 1999) and alter the decomposition rate of leaves after they are shed from the plant (Findlay and Jones 1990). Because species vary in their sensitivity, O_3 can shift the competitive balance in plant communities to the detriment of sensitive species (Miller and McBride 1999). Further, because individuals of a given species vary in their sensitivity, O_3 exposure can cause changes in genetic structure of populations, reducing or eliminating sensitive genotypes (Taylor *et al.* 1991; Davison & Reiling 1995).

At a broad scale, sensitivity of plants to O_3 is dependent on level of exposure, species, and soil-moisture status. The dependence on soil moisture reflects the fact that O_3 enters the plant through the stomates, and in dry conditions the stomates are more often closed. Thus, O_3 exposure in a dry year or at a dry site may be less damaging than the same exposure in more moist conditions (U.S. EPA 2006).

The complexity of the physical and chemical sources and sinks for atmospheric O_3 results in complex patterns of exposure in space and time. In lowland areas subject to air-pollution influence, O_3 concentration tends to increase during the day and reach a peak in late afternoon, then decline during the night to a minimum in early morning. On mountaintops this daily cycle may be absent, resulting in higher exposures for montane plants, especially in the morning hours (Aneja *et al.* 1994). Because O_3 reaches its greatest concentration downwind of, rather than within, major urban areas, O_3-sensitive trees may actually grow better in large cities than in the surrounding suburban and exurban areas (Gregg *et al.* 2003).

Many plants have been screened for O_3 sensitivity, but the screening is usually based on the development of visible foliar injury rather than on the more subtle responses of reduced photosynthesis or pathogen resistance. The U.S. National Park Service has compiled a list of plants in national parks that are known to be particularly sensitive to O_3 exposure (National Park Service 2003).

Despite the considerable research on plants, there is little information on the effects of O_3 on animals other than *Homo sapiens* and the animals that serve as its medical models, such as the Norway rat. Given the effects of O_3 on the human respiratory system, one might expect significant impacts of O_3 exposure on any animals with similar respiration mechanisms (Menzel 1984). However, we know of no information on O_3 effects on animals in natural ecosystems.

The O_3 research community has devoted considerable effort to synthesizing information on the effects of O_3 on plants. Several different indices of O_3 exposure are used, but one that is used commonly in plant research is the Sum06—the maximum, rolling 90-day sum of the average daytime [0800–1959 (8 A.M.–7:59 P.M.)] hourly concentrations of $O_3 \geq 0.06$ ppm for the year. A conference held to review O_3 exposure research and identify threshold levels of exposure that produce impacts on plants identified a Sum06 level of 8–12 ppm-hr as likely to produce foliar injury to some plants in natural ecosystems (Kohut 2007). Following the recommendation of the experts at that conference, 8–12 ppm-hr appears to be an appropriate red-line value for O_3 exposure. Because any O_3 can be injurious to sensitive plants (Fox *et al.* 1989), the best green-line value is probably the background, unpolluted level of O_3 exposure, but there is yet no consensus on what that background O_3 level was in the eastern United States.

Grasslands

Grasslands are a minor ecosystem type in the northeastern United States, and there is little information on the effects of air pollution on either the biota or the biogeochemistry of these ecosystems. Grasslands develop distinctly different communities, depending upon whether their soils are acidic or calcareous. Acid grasslands are more common, but calcareous grasslands tend to have more rare species (Stevens *et al.* 2006).

Nitrogen. We are aware of no direct studies on N effects on grasslands in the eastern United States However, experimental and gradient studies from elsewhere suggest that effects on species composition are likely (Dise and Stevens 2005). In Minnesota, N fertilization of an acid grassland (at a level of 100 kg N/ha-y) resulted in a 40% reduction in species richness over 12 years (Wedin and Tilman 1996). In Great Britain, a recent comparison of species composition in acid grasslands along a gradient of N deposition showed that species composition was affected at N deposition rates as low as 5 kg N/ha-y (Stevens *et al.* 2004). This deposition level is below the mean N deposition for the eastern United States, suggesting that current levels of N deposition are currently affecting grassland species composition in many areas. Thus, we have moderate confidence in the conclusion that N deposition is affecting acid grasslands in the eastern United States, and we suggest 5 kg N/ha-y as a preliminary green-line value, bearing in mind that no data are available from within the region. At the levels of deposition found in this region, the effects are likely to be shifts in relative abundance of species, favoring the nitrophilic species (such as *Agropyron repens,* Wedin and Tilman 1996), rather than loss of species or local extinction.

We know of no studies of N deposition on calcareous grasslands in the United States, Biogeochemically, they are likely to be less sensitive to acidification because the calcareous soils buffer the acidity. However, in Europe, species composition of calcareous grasslands is quite sensitive to N enrichment (Stevens *et al.* 2004). Species compositional shifts due to N enrichment may be more likely to cause species extinctions in calcareous grasslands because they tend to contain more rare species.

Sulfur, Ozone, and Mercury. We know of no studies of the effects of S, O_3, or Hg deposition on eastern U.S. grasslands. Because there are many plant and animal species that require grassland habitat, the lack of information on pollution effects is disquieting. Ozone is known to have effects on productivity of European grasslands (e.g., Bassin *et al.* 2007). In the case of Hg, however, grasslands are generally considered to have minimal abilities to methylate Hg, and data from a few studies indicate low Hg body burdens of grassland bird species (Evers *et al.* 2005).

Aquatic Ecosystems

The four air pollutants differ substantially in their effects on aquatic ecosystems. Sulfur is largely an agent of acidification through the mechanisms discussed earlier. Nitrogen can contribute to acidification but also can cause eutrophication (overenrichment with nutrients) in aquatic ecosystems that are limited by N supply. Mercury is a potent neurotoxin that accumulates in aquatic food webs to alter the behavior and reproduction of organisms at high trophic levels. Ozone has little effect in the water, but may have effects on emergent aquatic plants or air-breathing animals that are part of aquatic ecosystems. These potential effects of O_3 on aquatic plants and animals have not, to our knowledge, been studied, so we will ignore O_3 in the following discussion.

The effects of acid deposition on the chemistry and biology of aquatic ecosystems have been well known since the 1970s, and there is little doubt about the serious impact acidification has on a wide range of aquatic organisms (Weathers and Lovett 1998). The research has primarily focused on small lakes and streams and has included comparative studies across acid deposition gradients, experimental acidification of lakes and streams, and long-term studies of acidification or recovery in lakes or streams subject to increases or decreases in acid loading. Many of the key studies have been done in the target area of this synthesis (Northeastern and Mid-Atlantic regions of the United States). We have high confidence that acid deposition is adversely affecting aquatic ecosystems in this region.

Effects of acidic deposition on water quality include reduced pH (increased acidity), reduced acid-neutralizing capacity (ANC)[1], and increased aluminum (Al) concentrations (Driscoll *et al.* 2001). The primary variables of concern to organisms are pH and Al concentration. In gill-breathing organisms, Al interrupts gas and ion transport across respiratory membranes, leading to disruption of the five major functions of the gill: (1) ion transport, (2) osmoregulation, (3) acid–base balance, (4) N excretion, and (5) respiration (Brakke *et al.* 1994). The effects of Al on fish were known as early as the mid-1970s (Schofield 1978). Other physiological effects also occur in aquatic organisms, including altered hormonal and behavioral responses (Brakke *et al.* 1994). Aquatic organisms vary widely in their sensitivity to acidification. The most sensitive organisms tend to be adversely affected when

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[1] Acid neutralizing capacity (ANC) is the ability of water to neutralize strong acids, and is one of the primary measures of surface water acidification and recovery. Waters with ANC <0 μeq/L (micro-equivalents per liter) are considered chronically acidic, those with ANC between 0 and 50–100 μeq/L are considered sensitive to acidification, and those with ANC >100 μeq/L are often considered relatively resistant to acidification. While ANC does not directly affect aquatic organisms, it is an integrative measure for the propensity of an ecosystem to experience high acidity and Al concentrations, which do affect organisms.

TABLE 1. Sensitivity of Aquatic Taxa to Acidification

pH range	General biological effects
6.5 to 6.0	Little community change; possible effects on highly sensitive fish species (e.g., fathead minnow, striped bass)
6.0 to 5.5	Loss of sensitive species of minnows and dace (fathead minnow, blacknose dace). Perhaps decreased reproduction of walleye and lake trout; increased accumulation of filamentous green algae. Changes in species composition and decrease in species richness in phytoplankton, zooplankton, and benthic invertebrate communities. Loss of some zooplankton species and many species of clams, snails, mayflies, amphipods, and some crayfish
5.5 to 5.0	Loss of lake trout, walleye, rainbow trout, smallmouth bass, creek chub. Further increase in filamentous green algae. Loss of many zooplankton species as well as all snails, most clams, and many species of mayflies, stoneflies, and other benthic invertebrates
5.0 to 4.5	Loss of most fish species. Further decline in the biomass and species richness of zooplankton and benthic invertebrate communities. Loss of all clams and many insects and crustaceans. Reproductive failure of some acid-sensitive amphibians, including spotted salamanders, Jefferson salamanders, and the leopard frog

Source: Modified from Baker *et al.* 1990.

pH decreases below about 6, while some tolerant organisms can survive in waters as acid as pH 4. The sensitivity of various taxonomic groups to acidification has been well researched and some general patterns have been observed (Table 1).

Note that stream and lake acidification can be chronic or episodic, with episodic acidification usually occurring during large water-flow events such as large storms or snowmelt periods. The chemistry may be different in chronic vs. episodic acidification—for example, in the Northeast, N leaching is more important in episodic than in chronic acidification (Murdoch and Stoddard 1993; Wigington *et al.* 1996; Driscoll *et al.* 2001). Nonetheless, episodes of acidification can be as damaging to aquatic biota as chronic acidity, because a single event can kill an organism.

In addition to the direct toxic effects of pH and Al on aquatic organisms, indirect effects in lake and stream ecosystems can be important. For instance, lowered pH can reduce the concentration of dissolved organic carbon (DOC) in lakes (Effler *et al.* 1985; Monteith *et al.* 2007), allowing light to penetrate further into the lake. This increases the light available for macrophytes and benthic algae that grow on lake bottoms, and in addition the increased visibility alters the relationship between predators and prey in the lake (Yan *et al.* 2008). Dissolved organic carbon is important for another reason: it complexes aluminum and makes it less toxic, so a decrease in DOC increases the toxicity of Al. Indirect food-web effects can also occur; for instance, if a predator is tolerant of acidity but its prey are not, the predator will not be able to survive in an acidified lake (Baker *et al.* 1990).

Research on atmospheric deposition effects in lakes and streams clearly show that certain characteristics make lakes more susceptible to inputs of strong acids (Stoddard *et al.* 1998). In general, streams and lakes at higher altitudes, with thin till depth, noncarbonate geology, associated wetlands, and low ANC are considered sensitive to acidification. Perched seepage lakes recharged by rainwater are also considered sensitive (Young and Stoddard, 1996). Other factors influencing sensitivity to acid deposition include the ability of watershed soils to retain sulfate and nitrate. While some S is retained through biotic immobilization in soils and vegetation, most retention is through adsorption of sulfate on iron and aluminum oxides in the soil, and is usually highest in unglaciated soils (Galloway *et al.* 1983). In contrast, N retention is largely biological and is most complete in watersheds with rapidly growing forests. Forest type and previous land-use or disturbance history can also influence N retention (Lovett *et al.* 2002; Goodale and Aber 2001). Hydrology is important in stream acidification because flow paths that route water directly to the stream and minimize contact with the soil reduce the capacity for neutralization (Chen *et al.* 1984). In addition, there is considerable biodiversity present in intermittent streams and ephemeral ponds, but little research has been done on the factors controlling acidification in these environments.

Streams and lakes are not homogeneous environments, rather they encompass a range of habitat types that can vary in their acidification. For instance, many streams increase in pH as water moves downstream and the area of the contributing watershed increases (Schofield and Driscoll 1987). Even in headwater streams, some sections may be influenced by seepage from well-buffered groundwater and may represent a refuge for fish in an otherwise acidified stream (Baldigo and Lawrence 2001). Similarly, some invertebrates in a lake may escape from acidified water by sheltering in the well-buffered sediments on the bottom of the lake. Thus spatial heterogeneity in aquatic ecosystems is important for both the tolerance of and recovery from acidification. Further, this heterogeneity indicates that behavioral responses of organisms (e.g., the propensity of fish to drift downstream or invertebrates to burrow into the sediments during acid episodes) can influence the tolerance of the biota to acidification.

Mercury is deposited to aquatic ecosystems and their watersheds primarily in the ionic form. However, exposure to mercury is primarily associated with methylmercury; therefore, those factors that influence Hg methylation also influence the extent to which systems are affected by Hg deposition (Driscoll *et al.* 2007). The methylation process often proceeds with the involvement of sulfate-reducing bacteria, so conditions conducive to these bacteria, such as high sulfate and low oxygen, also promote more accelerated rates of methylation (Wiener *et al.* 2003). Most methylation of Hg occurs in the anoxic sediments of lakes, streams, and wetlands. Levels of dissolved organic carbon also appear to influence Hg methylation, but these effects are not completely understood and are the subject of current research. Methylmercury production and availability are also dictated by hydrology; on water bodies where large areas of substrate undergo wetting and drying, methylmercury levels are elevated (Evers *et al.* 2007).

Mercury is particularly dangerous in aquatic food chains where biomagnification is common. Food-chain lengths are primarily dictated by zooplankton diversity and abundance (Chen and Folt 2005; Chen *et al.* 2005). Greater zooplankton diversity can result in an order-of-magnitude increase in methylmercury in higher-trophic-level organisms, while in lakes with algal blooms, a "biodilution" effect can occur whereby methylmercury levels are dampened (Chen *et al.* 2005).

The known biological effects of methylmercury are numerous and likely affect all major vertebrate taxa at individual, population, and potentially at metapopulation levels. Effects can be categorized as physiological, behavioral, and reproductive. They are relatively well described for fish-eating wildlife and increasingly so for fish (see Evers 2005). Although traditional emphasis of Hg in fish has been on exposure to determine human and ecological effects, recent efforts have increasingly been placed on direct effects of methylmercury on fish that include inhibition of normal growth and gonadal development (Friedman *et al.* 1996), predator avoidance (Webber and Haines 2003), and on reproduction (Hammerschmidt *et al.* 2002). There are few exposure and effects studies on herpetofauna, although Hg is sometimes considered among the possible causes of long-term and widespread declines in amphibian populations (Bank *et al.* 2006).

Research on Hg in birds has been more comprehensive and has included both laboratory and field studies. Bird species in which the effects of Hg are well known from laboratory

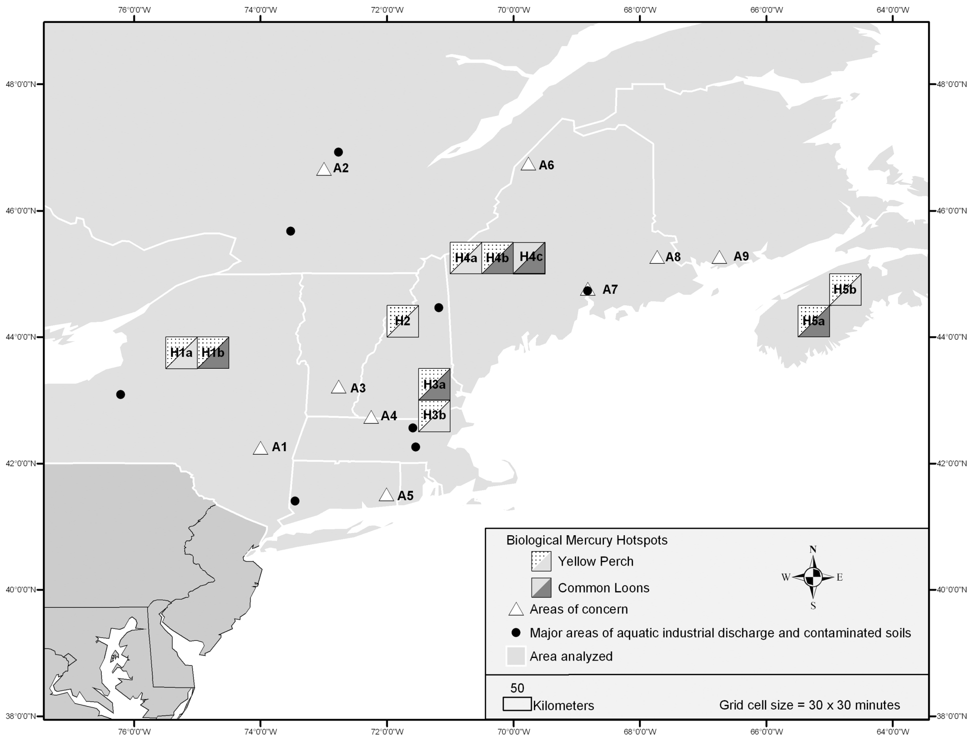

Figure 4. Biological mercury hotspots in the northeastern US and eastern Canada. Different shading patterns within the hotspot symbols reflect whether the hotspot was identified based on patterns in mercury concentration in perch or loons (the most commonly sampled species) or both. From Evers *et al.* 2007. Reproduced with permission of **Bioscience.**

studies include the mallard (*Anas platyrhychos*) (Heinz 1979) and ring-necked pheasant (*Phasianus colchicus*) (Fimreite 1971). Symptoms of mercury exposure in birds include decreases in reproductive success, behavioral changes such as a reduction in time spent hunting, and neurological problems such as brain lesions, spinal-cord degeneration, and tremors (Evers 2005). Until recently, results from Hg dosing studies on mallards have been used by the U.S. EPA (1997) and other agencies for setting universal threshold levels or lowest observed adverse-effect levels (LOAELs) for all bird species. Now, it is well understood that bird species vary in their sensitivities to methylmercury exposure. Sensitivity appears to be grouped by foraging guild: strict granivores appear to be most sensitive, while insectivores are more sensitive than omnivores, which are more sensitive than piscivores. Much of the effects literature is focused on piscivores, and for good reason, as piscivores have some of the most at-risk species. The species with the greatest literature on exposure and effects is the common loon (*Gavia immer*). Laboratory and field studies on both individuals and populations have developed national exposure profiles (Evers *et al.* 1998; 2003) as well as regional risk profiles (Evers *et al.* 2004) that are now related to "biological mercury hotspots" (Evers 2007) (Fig. 4). Lakes within these hot spots are now viewed as population sinks. Connectivity of these population sinks within the regional metapopulation of loons is currently being assessed. Other piscivorous birds in which Hg has been shown to harm reproductive success of populations include the bald eagle (*Haliaeetus leucocephalus*) in Maine (DeSorbo and Evers 2005), and wading birds such as the great egret (*Ardea alba*) in Florida (Bouton *et al.* 1999;

Spalding *et al.* 2000) and snowy egret (*Egretta thula*) in Nevada (Henny *et al.* 2002).

Recent studies suggest that the terrestrial invertebrate food web may have the ability to biomagnify Hg as much or more than the aquatic food web. Similar to the zooplankton food web, the transfer of methylmercury from one trophic level to the next (or from one secondary consumer to another) provides the ability for methylmercury concentrations to increase one order of magnitude. Spiders are predators in invertebrate food webs, and large spiders may carry as much of a methylmercury body burden as game fish. As a result, insectivorous birds such as songbirds and rails are likely at much higher risk than previously realized, even in strictly terrestrial habitats (Rimmer *et al.* 2005). Other more novel forage pathways for methylmercury are also coming to light and include food webs with periphyton and mollusks.

Mammal Hg exposure and effect levels, particularly for piscivorous species, are relatively well known. The best-studied species include mink (*Mustela vison*) and river otter (*Lontra canadensis*), for which sublethal effects include impairment of motor skills and weight loss. Yates *et al.* (2005) summarized exposure levels for the past two decades for much of northeastern North America. Laboratory studies on mink that establish LOAELs indicate effects in the wild are highly likely (Aulerich *et al.* 1974; Wren 1985, 1986, Dansereau *et al.* 1999).

With this background information in mind, in the next sections we summarize the effects of air pollution in various aquatic ecosystem types in the region of study.

High-Gradient Headwater Streams

Sulfur and Nitrogen. Sulfur and N deposition have two principal effects on headwater streams. First, if sulfate and nitrate leach through base-poor watershed soils, they can mobilize acidity and aluminum, which have direct effects on stream ecosystems. Second, N is often a limiting nutrient in aquatic ecosystems, and thus added N can produce eutrophication. In the past, headwater ecosystems in the temperate zone have usually been found to be limited by phosphorus, thus it has generally been assumed that the eutrophication effects are primarily expected in downstream ecosystems such as bays and estuaries that are more commonly N-limited. Recently, however, some headwater systems have been reported to be N-limited, and the eutrophication issue is currently being reassessed. (Bernhardt *et al.* 2005). With little information available, however, we will confine this discussion to acidification effects of sulfate and nitrate.

As stream acidity increases, sensitive species (see Table 1) will either die out or will seek refugia in less-acidified sections of the stream. Mobile species such as older fish and invertebrates will often "drift," essentially allowing themselves to be carried downstream in search of better habitat conditions. If the increase in acidity is brief (e.g., in response to heavy rainfall or snowmelt), there may be little mortality of adult fish unless species are prevented from leaving the system. When streams become chronically acidic, fish species can be lost from the ecosystem. There is strong evidence from many studies in this region and elsewhere that acidification can result in loss of fish populations and decline in fish species diversity (Jenkins *et al.* 2007). Research at Shenandoah National Park in Virginia suggests that one fish species is lost for approximately every 21 μeq/L decrease in ANC (Sullivan *et al.* 2002).

Effects on benthic species such as mussels and snails are not well studied. Naturally acidic headwater streams at high elevation do not normally support many mussel species. In episodically acidic streams mussels may close up to avoid toxic effects. Mussels would likely be eliminated from chronically acidic streams.

Mercury. Only recently has methylmercury in biota been measured in headwater streams. Salamanders have shown elevated Hg levels (Bank *et al.* 2005), and absence of some salamander species has been linked to potential chemical changes such as greater acidification and increased methylmercury availability (Bank *et al.* 2006). Further work has shown that crayfish in low-order streams generally have higher Hg burdens than crayfish in rivers, lakes, and reservoirs within the same watershed (Pennuto *et al.* 2005). There is compelling evidence that the high input of Hg and S in the Appalachian Mountains could have negative population-level impacts on the high diversity of salamander species that reside in upper watershed streams and ponds. (Bank *et al.* 2006)

Lakes and Ponds

While this synthesis includes both the lake and pond-rich glaciated regions of the northeastern United States and the unglaciated, lake and pond depauperate regions of the Mid-Atlantic, this section focuses primarily on lakes in the Northeastern region, as insufficient information is available for the small number of natural lakes in the Mid-Atlantic region. Atmospherically deposited S and N are both significant contaminants to this ecosystem. Mercury is also a significant contaminant, affecting a number of taxa across trophic levels.

Sulfur and Nitrogen. We have very high confidence that acidic deposition is affecting the biota of lake and pond ecosystems in the study area, particularly in the Northeast. Comparative studies of high- and low-acidity lakes within the region and experimental lake acidification studies elsewhere provide a scientifically coherent understanding of the effects of acidification (Schindler *et al.* 1985; Charles 1991). Animal species differ in their sensitivity to acidification (Table 1), but in general as pH decreases below 6 or ANC decreases below 100 μeq/L, taxa are progressively lost. In the Adirondacks, one fish species is lost for approximately every 21 μeq/L decline in ANC below 100 μeq/L (Fig. 5) (Sullivan *et al.* 2006). In lakes of the Adirondacks, an average of one zooplankton species is lost approximately with each 11 μeq/L decline in ANC (Sullivan *et al.* 2006). Remaining species may suffer directly from the effects of acidification, but also indirectly if some important food sources disappear as a result of acid stress, or species may respond positively if their predators are sensitive to acidity. In fact, the experimental removal of fish from unacidified lakes brings about some of the same changes that occur with acidification—invertebrate predators like corixids and *Chaoborus* become abundant along with a concomitant change in species composition and a decrease in biodiversity (Eriksson *et al.* 1980). Increased acidity is also linked to increased water clarity and, consequently, increased light penetration. This results in warming of the water column and altered stratification and seasonal turnover regimes.

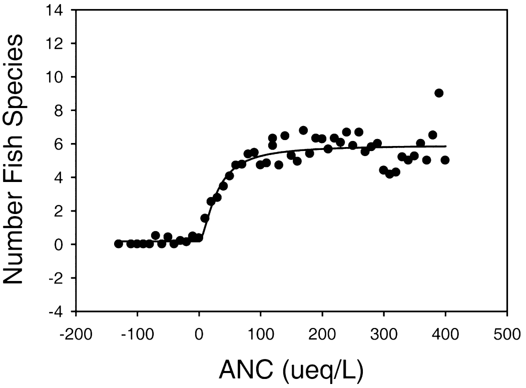

Figure 5. Number of fish species per lake as a function of acid neutralizing capacity (ANC) in Adirondack lakes. The data are presented as mean of species richness for every 10 ueq/L ANC class, based on data collected by the Adirondack Lakes Survey Corporation (after Sullivan *et al.* 2006).

Establishing one contaminant deposition threshold for all lakes across a diverse landscape is difficult, since there is a range of variability in natural ANC, pH, and base cation supply. Several available models, such as MAGIC (Cosby *et al.* 1985) and PnET-BGC (Gbondo-Tugbawa and Driscoll 2003) predict stream and lake acidification based on deposition levels and watershed characteristics. The parameter-intensive nature of these models has allowed their application to only a limited number of watersheds in the region. However, Cosby and Driscoll (in press) produced a map of sensitivity of aquatic ecosystems to acidification based on geologic characteristics in the unglaciated areas and on patterns of measured ANC in the glaciated region (Fig. 6).

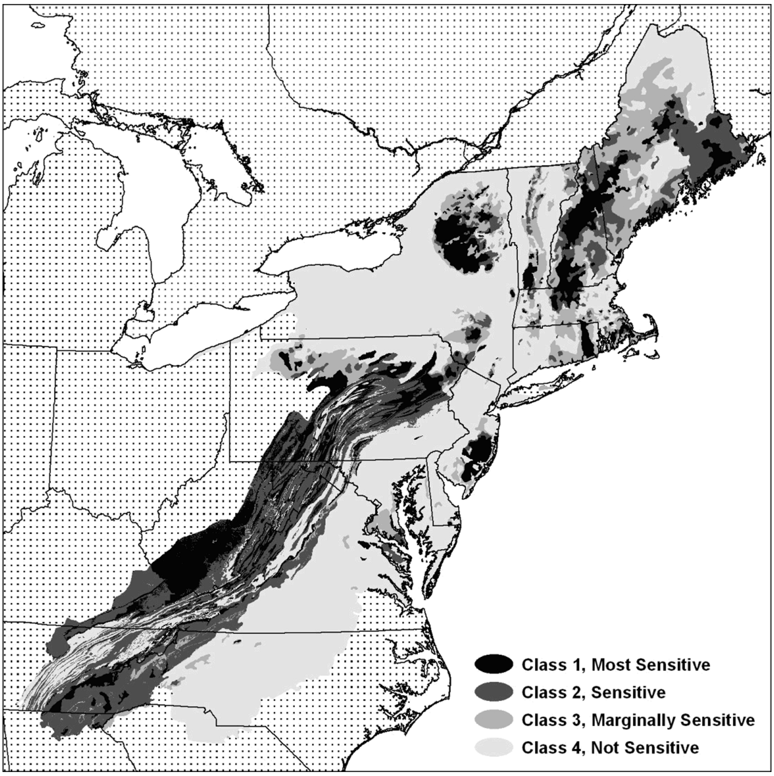

Figure 6. Map of landscape sensitivity to acidic deposition for the Northeastern and Mid-Atlantic Divisions of The Nature Conservancy. Four sensitivity classes are given based on an integrated assessment of the responses of a number of conservation targets in each class. Stippled areas were not considered in developing this map. (After Cosby & Driscoll in press.)

Atmospherically deposited N may have a eutrophying effect in addition to an acidifying effect. Export of N to estuaries can have important consequences, as discussed later in the chapter. Nitrogen is not usually a limiting nutrient in lakes and ponds (Driscoll *et al.* 2003); however, there is some evidence that very low-N surface waters may be limited or colimited by N, and therefore respond to N additions. Examples are streams in the New Jersey Pinelands (Morgan and Philipp 1986) and coastal plain ponds on Cape Cod (Kniffen *et al.* 2007) and possibly on Long Island as well.

Mercury. Atmospheric deposition, directly on the lake surface as well as on the watershed, is a significant source of Hg in lakes. A recent synthesis of Hg studies from the Northeast showed that Hg concentrations in fish tend to decrease with increasing pH, ANC, SO_4^{2-}, and total phosphorus and decreasing DOC in lakes (Chen *et al.* 2005; Driscoll *et al.* 2007). Land use also affects the sensitivity of lakes to Hg deposition, with a tendency for lower Hg levels in fish from aquatic ecosystems with higher residential and urban development in the watershed (Chen *et al.* 2005).

Much work has been conducted on piscivores residing in lakes, including the summary of large Hg exposure data for fish (Kamman *et al.* 2005), birds (Evers *et al.* 2005), and mammals (Yates *et al.* 2005) for northeastern North America. These efforts have provided insight into what species are at greatest risk. Fish species with the higher Hg levels include primarily introduced species such as walleye, northern pike, and both largemouth and smallmouth bass. Other species with elevated levels were yellow and white perch and lake trout. Although actual impacts from Hg on native fish species in lakes of the Northeast may be of lower concern than the ability of fish to transfer methylmercury to people, the known and potential impacts to fish-eating birds and mammals is of high conservation concern. There are documented impacts from methylmercury on the reproductive success of several piscivorous bird species, including the common loon and the bald eagle (Evers *et al*. 2005). Other species of concern based on compilations of Hg exposure levels and their piscivorous diet include the belted kingfisher, great blue and green heron, and common and hooded mergansers and mammals such as the mink and river otter (Evers *et al.* 2005; Yates *et al.* 2005).

We have a high degree of confidence that Hg has significant impacts on aquatic biota in the study area. Because the bioavailability of Hg depends on the extent to which it is methylated, and there is a wide variation in methylation abilities among lakes of different types, it is difficult to determine a threshold of Hg deposition that affects biota. Even relatively low levels of Hg deposition may have significant impacts if methylation rate is high and biomagnification occurs in the food web. Current modeling efforts will hopefully provide an ability to predict the propensity of Hg methylation in watersheds.

Low-Gradient Rivers

Sulfur and Nitrogen. Larger, low-gradient, lower-elevation streams and rivers in the Northeast and Mid-Atlantic do not generally show impacts from acid deposition. In larger watersheds, effects of atmospheric deposition are buffered by in-stream processes and the neutralizing capacity of the watershed. However, rivers may be sensitive to N pollution, especially as they near the sea and become estuaries or tidal rivers. We discuss the issue below under "Estuaries, Bays, and Salt Marshes."

Mercury The forested floodplains of large rivers have substantial ability to methylate Hg, and recent work at three Hg-contaminated rivers in the region (the Sudbury River in Massachusetts and branches of the Shenandoah and Holston Rivers in Virginia) has demonstrated the importance of floodplain forests and riparian wetlands for generation of high methylmercury levels (e.g., Waldron *et al.* 2000). Insectivorous bird species that live in these floodplains, such as the Carolina wren, worm-eating and yellow-throated warblers, Northern and Louisiana waterthrushes, red-winged blackbird, and song and swamp sparrows, have blood Hg levels that well exceed known toxic levels for songbirds (Evers *et al.* 2005). River Hg can also be transferred to the adjacent terrestrial food webs, including predatory spiders and the birds that feed on them (Cristol *et al.* 2008).

Estuaries, Bays, and Salt Marshes

Sulfur and Nitrogen. Estuaries, bays, and salt marshes are generally not severely impacted by acidic deposition, but they are subject to eutrophication caused by excess loading of N from atmospheric deposition and other sources (Boyer *et al.* 2002). These ecosystems are usually directly adjacent to, or receive water from, large rivers where there are high levels of N (and other nutrients) from a diversity of point and nonpoint sources, including agricultural and urban runoff, industrial and municipal wastewater, and atmospheric deposition to the estuary and to its watershed. The mix of these sources is unique to each watershed, but typically atmospheric deposition accounts for 25–40% of the total (Boyer *et al.* 2002; Castro and Driscoll 2002). Atmospheric deposition is often the largest single source of N to these ecosystems, though it contributes less than half of the total N loading. However, point sources and agricultural runoff of N are generally decreasing as better controls are put in place, while atmospheric N deposition is showing smaller declines (Driscoll *et al.* 2003). Thus, the percentage contribution of atmospheric deposition to this problem is likely increasing over time.

Most estuaries and bays in this region have some degree of eutrophication due to excess N loading (Scavia and Bricker 2006). The eutrophication leads to excess algal growth, and when the algae die and decompose, low oxygen concentrations may result, especially in deeper waters (Driscoll *et al.* 2003). The low oxygen is a danger to fish and shellfish, among other organisms (Scavia and Bricker 2006).

Salt marshes are well known for their ability to retain inorganic N, often responding with higher plant growth (Valiela *et al.* 1975). High plant growth and large accumulations of litter allow for substantial immobilization of N directly in plant tissue, microbial biomass, and organic matter accumulating during detritus decay (Findlay *et al.* 2002). A significant amount of research, much of it in the study area, has shown that nutrient additions to salt marshes can change species composition, generally allowing tall-form *Spartina alterniflora* to expand in coverage at the expense of higher marsh species, and that the plant-species change has measurable effects on animal consumers. (Sarda *et al.* 1996; Levine *et al.* 1998; Emery *et al.* 2001). These experiments typically involve levels of N deposition well above typical loading rates, so extrapolating to ambient conditions is difficult. Nonetheless, if the ecosystems are N limited, even small increments in N loading will probably have effects, perhaps subtle, on production and species composition. Comparative studies of salt marshes in Rhode Island show a negative relationship between N loading (much of the variation is driven by wastewater effluent loads) and plant-species richness such that over a range of watershed N loadings (∼1 to 30 kg N/ha marsh area-yr) species richness declines from roughly 10 to 5 species/transect (Wigand *et al.* 2003). The relationship is confounded to some degree by a covariation between N load and marsh physical characteristics.

Submerged aquatic vegetation (SAV) is known to be very sensitive to water quality with well-documented effects of eutrophication on water clarity and growth of epiphytes (Dennison *et al.* 1993; Stevenson *et al.* 1993). SAV supports a diversity of invertebrates and fishes, and is almost always a key variable in assessments of estuarine "condition" or "health." There have been several attempts to determine critical levels from either cross-system comparisons of N loads and SAV extent (e.g., Short and Burdick 1996). Values known to cause shifts in species composition (usually toward a macroalgal-dominated system) occur at loadings of about 25 kg N/ha-y and possibly less (see Hauxwell *et al.* 2003). These loadings are probably severalfold higher than loadings 100 years ago, and this N is derived from multiple sources with a predominance of wastewater (Roman *et al.* 2000).

Mercury It is well known that freshwater wetlands generally serve as areas of high Hg methylation, thus making obligate birds especially vulnerable to high levels of Hg contamination (Evers *et al.* 2005). The role of salt-marsh habitats in methylating Hg and enhancing its bioavailability (Marvin-DiPasquale *et al.* 2003), however, is less well documented, but is of increasing concern, especially in urban areas. Salt-marsh sharp-tailed sparrows are obligate salt-marsh passerines, with ≈ 95% of their global population breeding within the Northeast. Spending their entire annual cycle in salt-marsh habitats makes them excellent indicators of Hg contamination. Salt-marsh sharp-tailed sparrow blood Hg concentrations tend to be higher than other songbirds (Lane and Evers 2006). It is likely that salt-marsh sharp-tailed sparrows have significantly higher blood Hg levels because they feed at a higher trophic level or consume different prey than sympatric Nelson's sparrows (Shriver *et al.* 2006).

Synthesis and Linkages

In the previous section, we summarized the known effects of air pollution, ecosystem type by ecosystem type and pollutant by pollutant. While this approach allows us to focus on specific ecosystems and the conservation targets that inhabit them, it obscures issues and themes that cut across ecosystem boundaries and pollutant types. In this section we explore some integrative themes that emerge from this synthesis.

Intersystem Transfer of Pollutants

Many of the pollutants we discussed here are mobile and can be transported through a landscape, most often by drainage water. This has two principal consequences: (1) an atom of pollutant (S, N, or Hg) can have multiple effects as it is transported from ecosystem to ecosystem, and (2) "upstream" ecosystems can alter the rate, timing, and form of pollutant inputs to "downstream" ecosystems. We cite here several examples of these phenomena.

The Nitrogen Cascade

An atom of N released from fossil-fuel combustion can have multiple effects as it works its way through the environment. First, while in the atmosphere, it can contribute to the formation of O_3 and photochemical smog, harming both human health and ecosystem function. Next, if it is deposited to a terrestrial ecosystem, it can contribute to N saturation and its attendant effects—principally species compositional shifts and soil acidification. If the atom of N is denitrified in the soil and released to the atmosphere as nitrous oxide, it can contribute to the greenhouse effect. If, instead, it is leached through the soil into surface waters, it contributes to soil and stream acidification. Finally, as the N reaches estuaries and the coastal oceans, it can cause eutrophication, resulting in algal blooms, hypoxia, and other severe disruptions of the aquatic ecosystem. This series of effects of N in the environment has been termed the "nitrogen cascade" by Galloway *et al.* (2003), and is the reason why N pollution is so dangerous to the environment. It is also the reason why reduction of N emissions is so cost-effective in terms of environmental benefit per dollar spent in emission-control costs.

Mercury Methylation and Transfer

Throughout most of the eastern United States, Hg deposition has been enriched by a factor of 3 to 8 over values deposited a century ago (Lorey and Driscoll 1999; Kamman and Engstrom 2002). Elevated deposition of ionic Hg is converted to organic forms (by methylation) and incorporated in the food chain. The concentration of highly bioavailable methylmercury in surface waters is determined both by methylation in those surface waters and in the surrounding watershed. In particular, the presence of wetlands in the watershed increases the delivery of methylmercury to lakes because of the anaerobic soils in wetlands (Grigal 2002). Riparian zones are also important in the methylation of mercury (Driscoll *et al.* 2007). Thus Hg transformations in terrestrial wetlands and riparian zones can affect the rate of Hg bioaccumulation in a lake further downstream.

Aluminum in Soils and Surface Waters

As discussed earlier, deposition of S and N can acidify soils. Aluminum, an abundant natural component of soils, is in higher concentration in soil water from acidic soils because of its increased solubility in acidic solutions. Aluminum is toxic to roots because it can inhibit the uptake of nutrient cations such as calcium (Cronan and Grigal 1995). The soluble aluminum can also be leached into surface waters where it alters the osmoregulatory system of fish (Cronan and Schofield 1979; MacAvoy and Bulger 1995). Thus, a natural constituent of soils is mobilized by acid deposition and is transported downstream to be toxic in an ecosystem some distance from where it originally was mobilized. Moreover, the terrestrial ecosystem regulates the rate and timing of delivery of Al to surface waters by retaining or releasing sulfate and nitrate, the mobile anions that must accompany the Al. Changes in acid deposition rate may not be immediately reflected in proportional changes in Al leaching if S and N are stored in watershed soils and later released.

Interaction of Pollutants

Although we discussed pollutants individually in the second section, in reality they are all present simultaneously in most ecosystems in the eastern United States, and they can interact, sometimes in complex ways. These interactions can occur in the formation, deposition, or effects of the pollutants.

An example of an interaction effect on the formation of pollutants is the well-known role of N oxides in the photochemical reactions that form O_3. Thus the emission of one pollutant (nitric oxide) affects the formation of another (O_3). This process is complex and nonlinear, depending on the air temperature and the presence of hydrocarbons and sunlight (Warneck 1988).

An example of interaction in the deposition of pollutants is the role of ammonia in enhancing deposition of S. As sulfur dioxide gas is deposited to leaf surfaces, it acidifies those surfaces, which tends to slow down the sulfur dioxide deposition process. However, the alkaline nature of the ammonia gas (a pollutant gas emitted from agricultural operations and motor vehicle exhaust) counteracts the acidifying effect of the sulfur dioxide, leading to enhanced S deposition (Fowler *et al.* 2005).

There are many examples of the interaction of pollutants after they are deposited to ecosystems. Perhaps the simplest is the additive interaction of S and N deposition on soil acidification. Soil acidification is primarily driven by leaching of strong acid anions, which leaches the soil of basic cations such as calcium, magnesium, and potassium. Both sulfate and nitrate are anions that can cause acidification, and their effect is additive. [However, the situation becomes more complex if we try to predict the leaching of these ions based on their deposition rates, because of the many biological and abiotic processes that control the retention of deposited S and N in ecosystems (Fig. 2).]

Another example of the interaction of pollutants is interactive biogeochemical cycling of S and Hg. Deposition of sulfate stimulates the activity of a certain type of anaerobic bacteria that derive energy from the chemical reduction of sulfate. These same bacteria also methylate Hg, producing the form that accumulates in food chains. Thus the deposition of S stimulates the biogeochemical pathway that enhances the exposure of Hg. Experimental additions of sulfate to wetlands or lakes (Jeremiason *et al.* 2006; Branfireun *et al.* 1999) and long-term studies (Drevnick *et al.* 2007) show that changes in sulfate loading alter the formation of methylmercury and can alter concentrations of Hg in fish.

A third example of the interaction of pollutants within ecosystems involves the physiological responses of trees. Though not considered here, carbon dioxide is also a combustion-derived air pollutant. Increased concentrations of carbon dioxide tend to increase plant productivity, while O_3 usually decreases productivity. When they occur together, these responses can offset one another (Karnosky *et al.* 2003), thus either decreasing the phytotoxicity of O_3 or decreasing the growth response to carbon dioxide, depending at how one views the problem.

In general, much less research has been done on the interaction of pollutants than on the effects of single pollutants. The interactions are often complex, nonlinear, and poorly understood. Because these pollutants usually occur together, our understanding of the effects of air pollution will not be complete until we understand their interactions in much more detail and can predict their consequences.

Time Lags and Legacies

Ecosystems are complex amalgamations of biotic and abiotic materials, some of which respond quickly to environmental change and some very slowly. If air pollution affects one of the slowly changing components of an ecosystem, the pollutant effect is likely to endure for a long time. This seems obvious, but it bears some elaboration in examples that illustrate the ubiquity and timescale of these effects. For biotic effects, the issue of life span is critical. Effects of air pollution have primarily been demonstrated on short-lived organisms (e.g., aquatic invertebrates, mycorrhizae, grasses) and are much harder to demonstrate on long-lived organisms such as trees. Nonetheless, if air pollution does produce changes in tree-species composition, it may take centuries for the ecosystem to recover, because of the long generation time of trees. A similar example is the accumulation of S, N, and Hg in the organic matter of soils and lake sediments, which can have very slow decomposition rates. The accumulated S and N can leach out slowly from the soils over many decades after the cessation of the pollutant input. Recovery from base-cation depletion in soils can also be a very long-term process if the geologic substrate is low in these cations. If the pool of exchangeable cations has been depleted by years of acid deposition, the recovery of that pool requires inputs from atmospheric deposition and rock weathering that exceed outputs from leaching and accumulation in plants. The net rates are often low compared to the total pool, so this recovery can take decades or centuries (Driscoll *et al.* 2001).

Aquatic ecosystems are also subject to lag effects. A prime example is the recovery of biota in acidified surface waters. If acidification has caused local extinction of fish or invertebrates in a pond, and if reduction of pollutant deposition causes chemical recovery of the water quality, there may still be a time lag in the biotic response because dispersal and recovery of the organisms can be slow (Driscoll *et al.* 2001). Before a planktivorous fish population can be reestablished in a formerly acidified pond, first the chemical quality of the water must improve, then the invertebrates that the fish consumes must disperse to and reestablish in the pond, and finally the fish population itself must recolonize the pond in sufficient numbers to ensure a viable population. All of these steps can take time. This problem is compounded by the fact that as acid deposition is reduced, acidification of surface waters tends to become less chronic and more episodic, but often a single acid episode can disrupt biotic communities and require the recovery process to start from the beginning again.

Food-Web Effects

The preceding discussion leads to consideration of how food webs can control the expression of, and recovery from, pollutant effects. In lakes, for example, if a particular invertebrate species is acid-sensitive, the loss of that species as a lake acidifies will have ramifications not only for that species but also for the species that consume it, and likewise through the links of the food web. In addition to direct pollution effects, the biological effects of acidification include altered predator–prey interactions following the decline and disappearance of fish.

Bioaccumulation of Hg also depends strongly on food webs. Mercury toxicity is most evident in animals of high trophic levels, because Hg concentrations increase with each step up the food chain. Similar food-web effects may occur in terrestrial ecosystems, though they are less well documented. For example, the primary hypothesis for the effects of acid deposition on terrestrial birds is that soil acidification can reduce the abundance of ground-dwelling invertebrates that some birds require for adequate calcium supply (Hames *et al.* 2002).

Interactions Between Air Pollution and Other Environmental Changes

Just as the various pollutants that we considered interact with one another, their effects also interact with those of other environmental changes that are happening concurrently. There are myriad examples. Changing climate affects every biological and chemical process in ecosystems, from respiration of the leaves at the top of a forest canopy to microbial methylation of Hg in the sediment at the bottom of a lake. Microbial processes and activity of poikilothermic (cold-blooded) organisms are expected to be especially sensitive. Examples include the increase in bacterial nitrification with increasing temperature (Murdoch *et al.* 1998) and the effect of warming temperatures on the seasonal onset of calling in various species of frogs

(Gibbs and Breisch 2001). Temperature also effects the duration of ice cover on lakes (Likens 2000), the frost-hardiness of spruce trees subject to acid deposition (DeHayes *et al.* 1999), and many other aspects of ecosystem function. Temperature and moisture strongly affect the distribution of organisms (e.g., Iverson and Prasad 2001), so the whole biotic assemblage of ecosystems can change as climate shifts. Consequently, it is extremely difficult to predict the effects of pollutants on ecosystems, and the recovery of those ecosystems from reduced pollution, against the background of a changing climate.

Invasion of exotic species also interacts with air pollution. Deposition of N may make some habitats more suitable for weedy invasive plants (Howard *et al.* 2004; Jordan *et al.* 1997), and may make trees more susceptible to exotic pests (e.g., Latty *et al.* 2003). Invasive aquatic species can radically change the community composition of surface waters, thus altering the effects of acid deposition on the biota.

Land-use changes also influence the effects of air pollution, both by changing the distribution of emission sources and by changing the nature of the receiving ecosystems. Again, there are many examples, and we only list a few here. Most forest ecosystems in the northeastern United States are in a phase of regrowth from a period of heavy clearing in the nineteenth and early twentieth centuries (Foster *et al.* 1998). Their successional state strongly affects their retention and processing of N and their species composition (Aber and Driscoll 1997). Harvesting of timber and acid deposition both deplete crucial base cations from forest soils (Federer *et al.* 1989). Land-use changes in watersheds affect pollutant loading to aquatic ecosystems; for example, N from atmospheric deposition, agricultural runoff, and sewage treatment facilities all contribute to the eutrophication of estuaries (Fisher and Oppenheimer 1991). Increases in nutrient loading associated with urban and agricultural activities enhance the productivity of receiving waters but also may decrease fish Hg concentrations due to the biodilution phenomenon (Chen *et al.* 2005; Driscoll *et al.* 2007).

In general, while we may understand the effects of pollutants from controlled studies, the concurrent imposition of multiple forms of environmental change—air pollution, climate change, land use change, and exotic species—makes prediction of the responses of ecosystems to changing pollutant loading very challenging.

Conclusions and Implications

Air Pollution Has Significant Impacts on the Biodiversity and Functioning of Many Ecosystem Types

Effects of air pollution are known or likely to occur in all the ecosystems examined (Table 2). Thus, none of these ecosystem types is free of the impacts of air pollution, and most are affected by multiple pollutants. We do not yet understand all of the impacts of air pollution on ecosystems, but the available evidence indicates that air pollution is having a serious impact on the biodiversity and function of natural ecosystems across the eastern United States. In aquatic ecosystems, effects of acidity, N, and Hg on organisms and biogeochemical processes are well documented. Air pollution causes or contributes to acidification of lakes, eutrophication of estuaries and coastal waters, and Hg bioaccumulation in aquatic food webs. In terrestrial ecosystems, the effects of air pollution on biogeochemical cycling are well documented, and the effects on species composition are less well understood. Nevertheless, there is strong evidence for effects of N deposition on plants in grasslands, alpine areas, and bogs, and for N effects on forest mycorrhizae. Soil acidification is known to be occurring in some northeastern ecosystems, and is likely to affect the composition and function of forests in acid-sensitive areas over the long term. Ozone is known to produce reductions in photosynthesis in many terrestrial plant species.

TABLE 2. Level of Certainty that Air Pollutants Result in Significant Negative Impacts on Conservation Target Groups Discussed in This Review[a]

	Air pollutants and their products			
Conservation target groups	Nitrogen	Sulfur	Ozone	Mercury
Alpine and subalpine ecosystems	Likely	Likely	Unknown	Unknown
Forests (both upland and wetland types)	Likely	Known	Known	Likely
Bogs and fens	Likely	Known	Likely	Likely
Grasslands	Likely	Unknown	Unknown	Unknown
High-gradient headwater streams	Known	Known	Unlikely	Likely
Lakes and ponds	Known	Known	Unlikely	Known
Low-gradient rivers	Likely[b]	Unlikely	Unlikely	Likely
Estuaries, bays, and salt marshes	Likely[b]	Unlikely	Unlikely	Likely
Percent (number) of target groups with known or likely impacts	100% (8)	63% (5)	25% (2)	75% (6)

[a] Level of certainty was divided into four categories for ease of comparison across target and pollutant groups: known, likely, unlikely, and unknown. Known = studies documenting impacts in the region are known. Likely = studies documenting impacts are known, but none documented for this region; and/or plausible mechanism for impacts identified, but no specific studies to confirm the plausible link were identified. Unlikely = plausible links resulting in negative impacts are not supported at this time within or outside this region. Unknown = no applicable studies documenting impacts or lack of impacts were identified within or outside this region.

[b] Nitrogen eutrophication effects are known for these ecosystems. Although atmospheric nitrogen deposition is often a significant contributor to the total nitrogen loading in these ecosystems, there are also many other sources of nitrogen.

Air Pollution Impacts May Be Subtle, but Are Important

For the most part, the effects of these pollutants are chronic, not acute, at the exposure levels common in the Northeast. Mortality is often observed only at experimentally elevated exposure levels or in combination with other stresses such as drought, freezing, or pathogens. The notable exception is the acid/aluminum effect on aquatic organisms, which can be lethal at levels of acidity observed in many surface waters in the region.

The effects of these pollutants are subtle, but they can be serious. Changes in plant-species composition due to N enrichment may not cause immediate extinctions, but the effects can propagate through a food web to affect many organisms in an ecosystem. Likewise, increasing the N content of a tree may not kill it, but it may make it more susceptible to pests and pathogens that can kill it. Mercury may not kill fish but may reduce the reproductive success of the loons that eat them. Further, the effects of air pollution can interact with those resulting from other environmental changes, including climate change, land use change, and introduction of exotic species, to produce severe stress on natural ecosystems.

Critical Loads Are Important but Often Difficult to Identify

One of the goals of this effort was to identify levels of pollution that could help identify areas at risk of environmental damage—the red-line and green-line values discussed earlier. While we are aware of the challenges with setting simple critical-load or exposure values for complex and heterogeneous ecosystems, we know that identifying critical loads would be of considerable value to the conservation community, providing a basis for mapping impacts and thus allowing focus on sensitive areas and specification of target loading levels (Porter *et al.* 2005). We identified these levels where we could, but unfortunately in many cases the lack of specific information rendered us unable to specify a particular value. Often we were able to say with high or moderate certainty that impacts of a pollutant are occurring, even though we did not have sufficient information to identify a critical exposure level. Further research and further analysis of existing data should be focused on determining the most reasonable values for critical-load and exposure levels.

There Are Major Gaps in Our Knowledge

This review revealed major gaps in knowledge that will require much future research to fill. Among these gaps are:

- the effects of S and N deposition on species composition of terrestrial ecosystems, especially forests;
- controls on Hg methylation in lakes, wetlands, and terrestrial ecosystems;
- atmospheric dry deposition rates of pollutants, especially Hg;
- exposure and effects of Hg on organisms in terrestrial ecosystems;
- base cation weathering rates in soils;
- effects of O_3 on grassland, wetland, and alpine plants and on wild animals;
- interactive effects of pollutants;
- factors controlling recovery rates from acidic deposition in lakes, streams, and soils.

Filling these knowledge gaps will require a sustained investment in research. Unfortunately, funding for air-pollution research (other than greenhouse gases) has declined precipitously since the 1980s, and shows no sign of increasing in the foreseeable future. This puts scientists and resource managers in a precarious situation of knowing enough to be very concerned about the effects of air pollution on natural ecosystems, knowing that there is much that we don't understand, and being unable to improve that situation for lack of funding.

One particular type of research is especially crucial and deserves special mention. The United States does not have an integrated environmental monitoring system. Without monitoring data, we are in many cases unable to determine if populations are declining, or, if pollution is reduced, if they are recovering. Chemical monitoring of precipitation and surface waters is sparse but has been used very effectively in policy formulation (Lovett *et al.* 2007). Biological monitoring is primarily an ad hoc activity by individual scientists who try to sustain the necessary long-term funding. Without a comprehensive, integrated environmental monitoring program, it is difficult to track the chemical and biological responses to air pollution.

Air-Pollution Impacts Are Most Likely Being Underestimated

It is apparent from this review that we currently have limited understanding of some potential effects of specific air pollutants (e.g., effects of N deposition on alpine ecosystems, effects of O_3 on wildlife), the strength of interactions among pollutants, the extent and implications of time lags, the relative amount of primary and secondary food-web effects, and the nature of interactions with other environmental threats. Unfortunately, there are many areas of research that have been largely discontinued (e.g., ground-level O_3 impacts to plants), and other areas of inquiry that illustrate impacts are much more pervasive than previously thought (e.g., soil acidification, Hg). Taken together, it seems likely that our current knowledge, as summarized in this chapter, represents an underestimation of the problem.

Implications for Conservation

The impacts of air pollution on humans and the natural world have been known for a long time (Weathers and Lovett 1998). However, in most cases these impacts have been studied as smaller pieces of a larger puzzle—such as a single pollutant's impact to an individual species or ecosystem. For effective conservation of biological diversity we must look across multiple species and ecosystems and assess the scope and severity of the threat presented by multiple pollutants at the same time. In this synthesis we attempt, for the first time to our knowledge, to look broadly at the impacts of air pollutants to those species, ecological communities, and ecosystems that are the focus of biodiversity conservation. Our assessment revealed that in the Northeast and Mid-Atlantic regions of the United States, the impacts are significant and widespread across many ecosystem types, disrupting the functioning of many of these ecosystems to varying extents, and harming species and communities of conservation concern. Conservation organizations have largely focused on habitat protection as the main tool for preserving important species and communities. However, preserving land will not protect these species and communities from threats such as air pollution that do not recognize property boundaries. We believe that conservation organizations should consider the impacts of air pollution in their conservation agendas, in order to protect the wildlife species at risk, the ecosystems that provide valuable services to our society, and the centuries of personal and financial investments made to preserve these ecosystems for humans and wildlife.

Recommendations for Policy and Science

Based on our evaluation of the science as summarized in the preceding sections, further action should be taken to help protect ecosystems and biodiversity from the damaging effects of air pollution. Two principal changes are most important at this time: (1) the nation should set ecologically relevant air-pollution standards, and (2) federal, state, and private organizations should work together to develop a comprehensive monitoring program for air pollution and its effects.

Set Ecologically Relevant Air-Pollution Standards

Current air-pollution regulations are based on the National Ambient Air Quality Standards (NAAQS), which are set for "criteria pollutants," including sulfur dioxide, nitrogen oxide, and ozone, among others. The primary NAAQS standards are set to protect human health, and although the Clean Air Act also allows for secondary standards that are intended to protect "public welfare," including buildings, crops, and natural ecosystems, in most cases secondary standards have simply been set equal to primary standards. The primary standards represent levels of concentration of the pollutant in the air which, if exceeded, are deemed to represent a significant threat to health. While necessary, these standards are insufficient to protect ecosystems from air pollution.

Ecosystems respond to different pollutants, different exposure indices, and over different timescales than human bodies. For S, N, and Hg, the most logical index of exposure of ecosystems is total annual deposition, such as kilograms of the pollutant deposited per hectare per year. Ecosystem response is best captured in a measure of total deposition of S and N, rather than concentrations of chemical species such as sulfur dioxide or nitrogen oxide. For Hg, there is much less information to assess the appropriate exposure index, but total annual deposition also appears to be a reasonable choice. For ozone, a cumulative exposure statistic such as Sum06 (see the earlier discussion) is probably a more relevant index of the effects on plants than is a mean or maximum concentration.

Deposition standards for S and N are most often expressed as critical loads. Critical loads have been used for assessing the impacts of S and N deposition in Europe for over two decades (Burns *et al.* 2008). We recommend that the U.S. adopt a critical-loads framework for assessing the effects of S and N deposition on sensitive ecosystems, and explore this approach for Hg as well. Critical loads should be viewed as evolving standards, and should be periodically updated based on the latest research, as is done for the criteria pollutants. Investment in research and scientific consensus-building will be required to develop an initial estimate of the critical load for each pollutant and ecosystem type. It may be prudent to establish an initial set of critical loads based on best available science that are tested with empirical data collected from priority areas with sensitive ecosystems. This will require improvements in current monitoring systems and a process to feed this information back into air pollution regulations.

Develop a Comprehensive Monitoring Program for Air Pollution and Its Effects

Despite years of research documenting the impacts of air pollution on wildlife and ecosystems, currently there is no integrated national monitoring program in place to measure the comprehensive effects of changing emissions. Monitoring of atmospheric deposition and surface-water chemistry has been essential to U.S. policy formulation and assessment, but the funding for these programs is constantly under threat. Further, there is no comprehensive monitoring of the impacts of air pollution on forests, soils, or most plants and animals. Therefore, we recommend that the federal and state governments and nongovernmental organizations such as universities and conservation organizations work together to create and integrated and comprehensive monitoring program for the natural resources at risk from air pollution. Such an initiative should include and expand current networks for measuring precipitation chemistry, air quality, and surface-water chemistry, and would include new programs to monitor forests, soils, wildlife, and other natural resources that are threatened by air pollution. A comprehensive and integrated monitoring network would help address important information gaps and inform the development and refinement of critical loads. The comprehensive monitoring program should be long-term and national in scope, and increased funding and commitment to long-term stability of the programs will be required. Without an effective monitoring program in place, we cannot evaluate the impacts of air pollution on ecosystems, nor can we determine if legislation intended to reduce these impacts is having the desired effects.

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Conflicts of Interest

The authors declare no conflicts of interest.

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