Effects of Atmospheric Deposition of Acidity and Mercury on Freshwaters and Other Sensitive Natural Resources in the Eastern U.S.

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EXECUTIVE SUMMARY

In this testimony, the ecological effects of atmospheric sulfur, nitrogen and mercury deposition to sensitive resources in the eastern U.S. are summarized. Acidic deposition is comprised of sulfuric and nitric acids, and ammonium derived from atmospheric emissions of sulfur dioxide, nitrogen oxides, and ammonia, respectively. These compounds are released to the atmosphere largely by the burning of fossil fuels and agricultural activities. Mercury emissions in the U.S. are largely the result of electric utilities, incinerators and industrial processes. Atmospheric deposition delivers acids and acidifying compounds and mercury to the Earth's surface. The adverse ecological effects of acidic and mercury deposition on forests, streams and lakes are summarized and the need for adequate monitoring to track the effectiveness of emission control programs on water resources is emphasized.

Forest Ecosystems

Acidic deposition has altered, and continues to alter, forest soil by accelerating the leaching of calcium and magnesium and increasing concentrations of dissolved inorganic aluminum in soil waters. At high concentrations, dissolved inorganic aluminum can hinder the uptake of water and essential nutrients by tree roots.

The alteration of soils by acid deposition has serious consequences for acid-sensitive forest ecosystems. Soils that are compromised by acidic deposition are less able to neutralize additional inputs of strong acids, and provide poorer growing conditions for plants and delay the recovery of surface waters.

Freshwater Aquatic Ecosystems

Acidic deposition has impaired, and continues to impair, the water quality of lakes and streams in the eastern U.S. in three important ways: lowering pH levels (i.e., increasing the acidity); decreasing acid-neutralizing capacity (ANC); and increasing aluminum concentrations. Many surface waters in New England, the Adirondack region of New York, and the Northern, Central and Southern Appalachian Mountain regions exhibit chronic and/or episodic (i.e., short-term) acidification. Moreover, elevated concentrations of dissolved inorganic aluminum have been measured in acid-impacted surface waters throughout the East.

High concentrations of aluminum and increased acidity have reduced the species diversity and abundance of aquatic life in many lakes and streams draining acid-sensitive regions in the East. Fish have received the most attention to date, but entire food webs are often negatively affected. For example, in a survey of lakes in the Adirondacks, 346 lakes (24% of the total) did not contain fish. These fishless lakes had significantly lower pH and higher concentrations of dissolved inorganic aluminum when compared to those lakes with fish.

There are important linkages between acidic deposition and other water quality problems. For example, mercury contamination of fish is coupled to surface water acidification through a pattern of increases in fish mercury concentration with decreases in surface water pH. Studies across the eastern U.S. have shown that many surface waters have elevated concentrations of mercury in fish tissue as a result of atmospheric emissions and deposition of mercury. "Biological mercury hotspots" have been identified at five areas in eastern North America.

Monitoring Ecosystem Response to Emission Controls

Environmental monitoring is a critical tool to help track the effectiveness of past controls of emissions of air pollutants and to guide future air quality management in the U.S. There are several national programs which are widely used by the research and policy communities to evaluate the extent and change in atmospheric deposition and to assess changes in surface water chemistry in response to changes in emissions of air pollutants. Unfortunately, there are two items in the President's 2008 budget that will substantially jeopardize these programs. This budget shows a $1 million cut to the Clean Air Status and Trends Network (CASTNet) program (which includes measurements of air chemistry and dry deposition of acidic substances) and a cut of $5.75 million (effectively zeroing out the program) to the extramural programs through which the status of acid-sensitive surface waters are monitored by the U.S. Environmental Protection Agency. Without these two critical monitoring programs it will be difficult if not impossible to track the response of atmospheric chemistry and acid-sensitive surface waters to current and future controls on emissions of air pollutants.

There is also a critical need to develop a national program for monitoring ecosystem response to controls on emissions of mercury to the atmosphere. This need was recently emphasized by the introduction of bills in both the House of Representatives and the Senate to establish such a program through a multi-agency initiative (House - H.R. 1533 "Comprehensive National Mercury Monitoring Establishment Act" Sponsor: Tom Allen (D-ME). Cosponsors: James Walsh (R-NY), John McHugh (NY), Michael Michaud (ME), Mark Steven Kirk (IL), Raul Grijavla (AZ); Senate - S. 843 "Comprehensive National Mercury Monitoring Act" Sponsor: Susan Collins (R-ME), Joseph Lieberman (R-CT), Hillary Clinton (D-NY)).

1.0 INTRODUCTION

Detailed studies by a large community of scientists for more than three decades have provided considerable insight into the ways in which atmospheric deposition alters ecosystems. When it was first identified (e.g. Gorham 1989), atmospheric deposition was viewed as a simple problem that was limited in scope. Scientists now know that acids and acidifying compounds and mercury largely enter ecosystems from atmospheric deposition and are transported through soil, vegetation, and surface waters, resulting in a range of adverse ecological effects.

In this testimony, I present information on patterns of acidic and mercury deposition, and the effects of atmospheric deposition of sulfur, nitrogen and mercury on sensitive forest, and freshwater resources. I also make recommendations to strengthen the national monitoring programs to track ecosystem response to air quality management.

2.0 ACIDIC DEPOSITION

Acidic deposition is largely comprised of sulfuric and nitric acid derived from sulfur dioxide and nitrogen oxides, respectively, and ammonium resulting from emissions of ammonia. Sulfur dioxide and nitrogen oxides, originating from human activities, are largely emitted to the atmosphere by the burning of fossil fuels, while ammonia is largely the result of agricultural activities. Once these compounds enter an ecosystem, they can acidify soil and surface waters, bringing about a series of ecological changes. The term acidic deposition encompasses all of the forms of these compounds that are transported from the atmosphere to the Earth, including gases, particles, rain, snow, clouds, and fog. Acidic deposition occurs as wet deposition: as rain, snow, sleet or hail; as dry deposition, as particles or vapor; or as cloud or fog deposition, which is more common at high elevations and in coastal areas. Wet deposition is fairly well characterized by monitoring at more than 200 National Atmospheric Deposition Programs (NADP) in the U.S. In contrast dry deposition is highly dependent on meteorological conditions and vegetation characteristics, which can vary markedly over short distances in complex terrains. As a result dry deposition is poorly characterized and highly uncertain. Dry deposition is characterized through the Clean Air Status and Trends Network (CASTNet), which include 97 sites in the U.S.

Sulfuric and nitric acids lower the pH of rain, snow, soil, lakes, and streams. In 2002-2004, wet deposition (i.e., deposition from forms of precipitation such as rain, snow, sleet, and hail) in acid-sensitive regions of the eastern U.S. had average pH values of 4.3 to 4.5, which is about three to five times more acidic than background conditions. Wet deposition of sulfate and nitrate are elevated in the eastern U.S. (Figure 1, 2).

Acidic deposition trends in the eastern U.S. mirror emission trends in the atmospheric source area, which extends to the Midwest (Lynch et al. 2000; Butler et al. 2001; Likens et al. 2000; Driscoll et al. 2001; Driscoll et al. 2003). Long-term data from across the eastern U.S. show declining concentrations of sulfate in wet deposition since the mid-1970s, coincident with decreases in sulfur dioxide emissions (Lynch et al. 2000; Driscoll

et al. 2001; Driscoll et al. 2003). Based on these long-term data, a strong positive correlation exists between sulfur dioxide emissions in the source area and sulfate concentrations in wet deposition (Figure 3; Butler et al. 2001; Driscoll et al. 2001). It is now expected that the sulfate concentration of wet deposition in the eastern U.S. will increase or decrease in a direct linear response to the increase or decrease of sulfur dioxide emissions in the atmospheric source area. These observations strongly suggest a cause and effect relationship between emissions of sulfur dioxide and deposition of sulfate in sensitive regions in the eastern U.S. A similar relationship is starting to become evident between emissions of nitrogen oxides and wet deposition of nitrate. This relationship for nitrate is not as strong as the relationship for sulfur because emissions of nitrogen oxides have been relatively constant over the last 20 years. However, it appears that recent decreases in emissions of nitrogen oxides from electric utilities are starting to drive reductions in atmospheric deposition of nitrate (Butler et al. 2003).

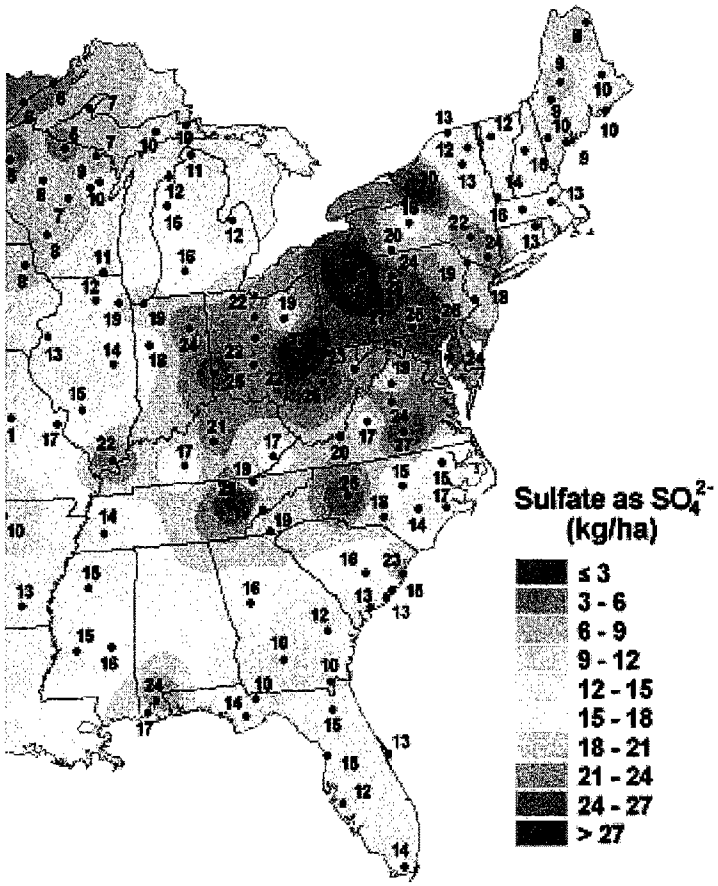

Figure 1. Map of wet sulfate deposition for the eastern U.S. for 2003. Data are from the NADP network.

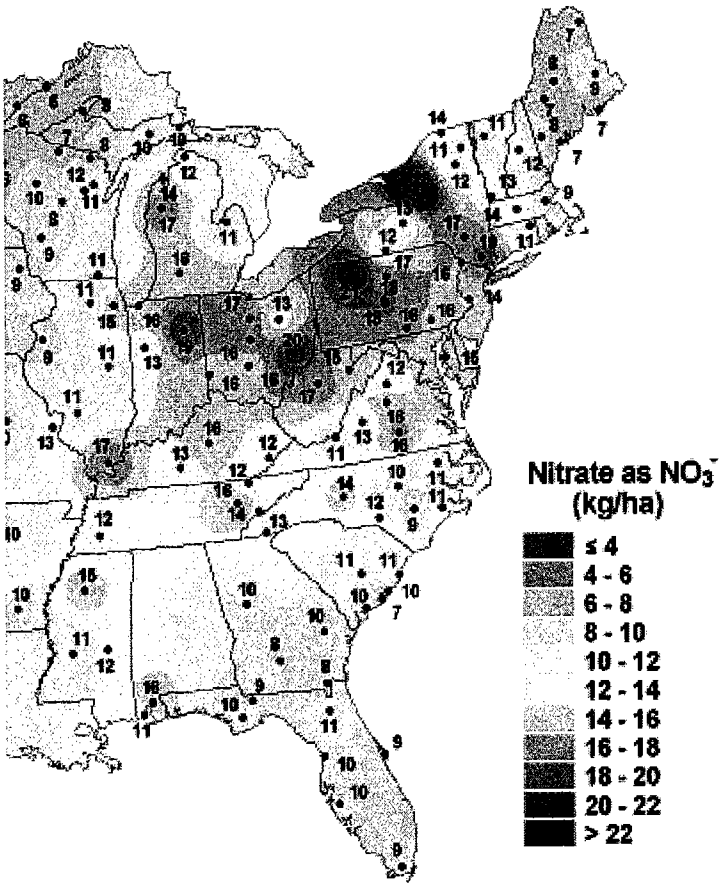

Figure 2. Map of wet nitrate deposition for the eastern U.S. for 2003. Data are from the NADP network.

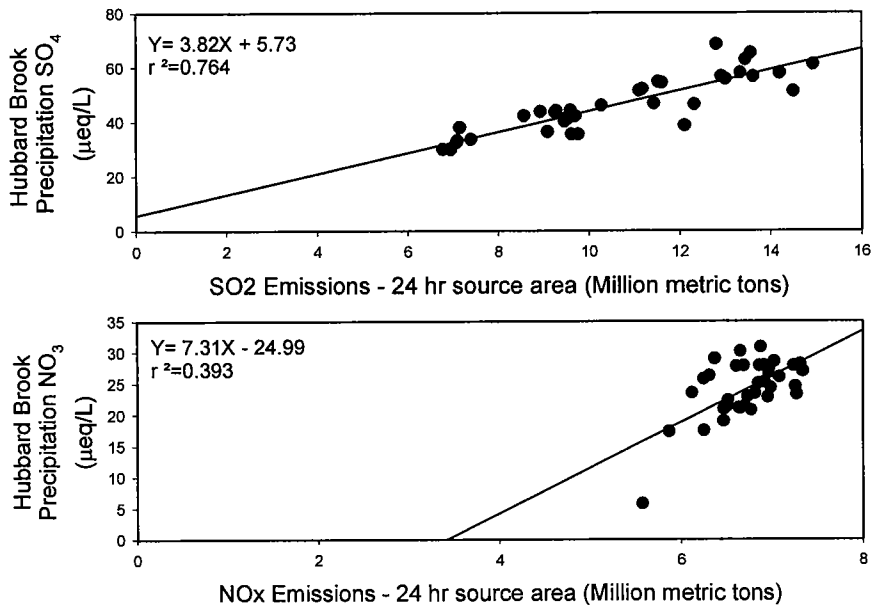

Figure 3. Volume-weighted annual concentrations of sulfate and nitrate in bulk precipitation at the Hubbard Brook Experimental Forest, New Hampshire as a function of annual emissions of sulfur dioxide and nitrogen oxides for the source area based on 21-hr back trajectory analysis.

3.0 Effects of Acidic Deposition on Forest and Aquatic Ecosystems of the Eastern U.S.

In acid-sensitive regions across the eastern U.S., acidic deposition alters soils, stresses forest vegetation, acidifies lakes and streams, and harms fish and other aquatic life (Figure 4; Driscoll et al. 2001). These effects can interfere with important ecosystem functions and services such as forest diversity and productivity and water quality. Years of acidic deposition have also made many ecosystems more sensitive to continuing pollution. Moreover, the same pollutants that cause acidic deposition contribute to a wide array of other important environmental issues at local, regional, and global scales (see Table 1).

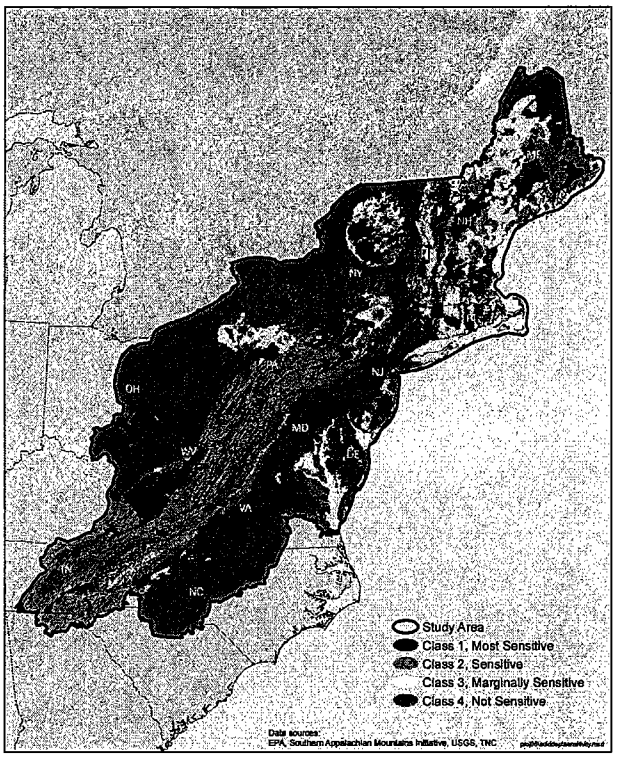

Figure 4. Map showing sensitivity of the eastern U.S. to acidic deposition. Four sensitivity classes are shown. Sensitivity class 1 represents waters with ANC values < 50 μeq/L. Sensitivity class 2 represents waters with ANC values 50-100 μeq/L. Sensitivity class 3 represents waters with ANC values 100-200 μeq/L. Sensitivity class 2 represents waters with ANC values >200 μeq/L.

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

Problem	Linkage to Acid Deposition	Reference
Coastal eutrophication	Atmospheric deposition adds nitrogen to coastal waters.	Jaworski et al. 1997; Paerl et al. 2000; Castro and Driscoll 2002
Mercury	Surface water acidification increases mercury accumulation in fish.	Grieb et al. 1990; Driscoll et al. 1994; Weiner et al. 2003
Visibility	Sulfate aerosols diminish visibility and views.	Malm et al. 1994
Tropospheric ozone	Emissions of nitrogen oxides contribute to the formation of ozone.	NAPAP 1998; Gunthardt-Goerg et al. 2000

3.1 Effects of Acidic Deposition on Forest Soils

Research has shown that acidic deposition has chemically altered soils with serious consequences for acid-sensitive ecosystems. Soils compromised by acidic deposition lose their ability to neutralize continuing inputs of strong acids, provide poorer growing conditions for plants, and extend the time needed for aquatic ecosystems to recover from acidic deposition.

Acidic deposition has altered and continues to alter soils in sensitive regions of the Eastern U.S. in three important ways. Acidic deposition depletes available calcium and other nutrient cations from exchange sites in soil; facilitates the mobilization of dissolved inorganic aluminum into soil water; and increases the accumulation of sulfur and nitrogen in soil.

Loss of calcium and other nutrient cations

The cycling of calcium and other nutrient cations in forest ecosystems involves the inputs and losses of these materials. For most forest ecosystems the supply of calcium and other nutrient cations largely occurs by weathering (i.e., the breakdown of rocks and minerals in soil). Calcium and other nutrient cations may also enter forests by atmospheric deposition, although this pathway is generally much smaller than weathering. Losses largely occur by vegetation uptake and drainage waters. An important pool of ecosystem calcium and nutrient cations is the soil available pool or the soil cation exchange complex. Plants are generally able to utilize this source of nutrients. Forest ecosystems that are naturally sensitive to acidic deposition are generally characterized by low rates of weathering and generally low quantities of available nutrient cations. Under conditions of elevated inputs of acidic deposition and subsequent transport of sulfate and nitrate in drainage waters, nutrient cations will be displaced from available pools and leached from soil (Reuss and Johnson 1986). This condition is not problematic for areas with high weathering rates and high pools of available nutrient cations. However, in acid-sensitive areas with shallow soil which contain minerals that are resistant to weathering, the enhanced loss of calcium and other nutrient cations can result in a depletion of soil available pools.

Over the last century, acidic deposition has accelerated the loss of large amounts of available calcium from acid-sensitive soil in acid-sensitive areas in the Northeast (Likens et al. 1996; 1998; Bailey et al. 1996; Lawrence et al. 1999). This conclusion is based on more than 20 studies throughout the eastern U.S. Depletion occurs when nutrient cations are displaced from the soil by acidic deposition at a rate faster than they can be replenished by the slow breakdown of rocks or the deposition of nutrient cations from the atmosphere. This depletion of nutrient cations fundamentally alters soil processes, compromises the nutrition of some trees, and hinders the capacity for sensitive soils to recover. Likens et al. (1996, 1998) observed that more that half of the available calcium had been lost from soil at the Hubbard Brook Experimental Forest, New Hampshire over the past 60 years. Marked decreases in exchangeable calcium and magnesium over the

past 20 years are evident from re-sampling of soils in the Northeast (Warby et al. in review).

Mobilization of aluminum

Aluminum is often released from soil to soil water, lakes, and streams in forested regions with high acidic deposition as well as low stores of available calcium and high acidity in soils (Cronan and Schofield 1990; Driscoll and Postek 1995). One of the most significant ecological effects of acidic deposition is the mobilization of aluminum from soil and a shift in the form of aluminum in water from non-toxic organic forms to highly toxic inorganic forms.

Concentrations of aluminum increase markedly with decreases in pH, particularly the toxic inorganic forms of aluminum. It is evident that concentrations of aluminum increase exponentially when lake pH decreases below 6. Aluminum concentrations are thought to be ecologically significant when they increase to values above 2 μmol/L (Driscoll et al. 2001). This condition clearly occurs below pH 6.0.

High concentrations of dissolved inorganic aluminum can be toxic to plants (Cronan and Grigal 1995), fish (MacAvoy and Bulger 1995), and other organisms. Concentrations of dissolved inorganic aluminum in streams in New York, New England and in the Appalachian Mountains are often above levels considered toxic to fish (McAvoy and Bulger 1995) and much greater than concentrations observed in forest watersheds that receive low inputs of acidic deposition (Driscoll et al. 1988).

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

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

4.0 Effects of Acidic Deposition on Freshwater Aquatic Ecosystems of the Eastern U.S.

4.1 Surface water acidification

Acidic deposition degrades surface water quality by lowering pH (i.e., increasing acidity); decreasing acid-neutralizing capacity (ANC); and increasing dissolved inorganic aluminum concentrations. While sulfate concentrations in lakes and streams have decreased in the northeastern U.S. over the last 20 years, they remain high compared to background conditions (e.g., approximately 20 μeq/L; Driscoll et al. 1991; Brakke et al. 1989; Chen and Driscoll 2005a,b). Moreover, improvement in other chemical conditions

in many lakes and streams in acid-impacted regions of the eastern U.S. has been limited (Likens et al. 1996; Stoddard et al. 1999; Driscoll et al. 2001; Stoddard et al. 2003).

For example, a comprehensive survey of lakes greater than 0.2 ha in surface area in the Adirondack region of New York was conducted by the Adirondack Lakes Survey Corporation to obtain detailed information on the acid-base status of waters in this region (Kretser et al. 1989). Of the 1469 lakes surveyed, 24% had summer pH values below 5.0. Also 27% of the lakes surveyed were chronically acidic (i.e., ANC less than 0 μeq/L) and an additional 21% were susceptible to episodic acidification (i.e., ANC between 0 and 50 μeq/L. An analysis of the anion content of these lakes illustrates that these lakes have predominantly been acidified by atmospheric deposition of sulfate. Other regions of the eastern U.S. showing impacts of acidic deposition on surface waters include New England, and the Northern, Central and Southern Appalachian Mountain regions (Charles 1991).

Seasonal acidification is the periodic increase in acidity and the corresponding decrease in pH and acid neutralizing capacity in streams and lakes (Wigington et al. 1996). Episodic acidification is caused by the sudden pulse of acids and a dilution of base cations (e.g., calcium, magnesium, sodium, potassium) due to spring snowmelt and large rain events in the spring and fall. Increases in nitrate are often important to the occurrence of acid episodes. These conditions tend to occur when trees are dormant and therefore using less nitrogen. At some sites, short-term increases in sulfate and organic acids can also contribute to episodic acidification. Episodic acidification often coincides with pulsed increases in concentrations of dissolved inorganic aluminum. Short-term increases in acid inputs to surface waters can reach levels that are lethal to fish and other aquatic organisms (Baker et al. 1996; Van Sickle et al. 1996). All of the acid-sensitive and acid-impacted regions discussed in this report (i.e., Adirondacks, Catskills, New England, Appalachian mountains) have documented effects associated with episodic acidification (Van Sickle et al. 1996; Webb 2004).

In summary, it is well documented that surface waters in New England, the Adirondacks, and the Northern, Central and Southern Appalachian mountain regions have been adversely impacted by elevated inputs of atmospheric sulfur and nitrogen deposition. Surface waters in these areas exhibit chronically acidic conditions or have low values of acid neutralizing capacity, which make them susceptible to short-term episodic acidification.

Regional trends in surface water chemistry indicate that recovery of sensitive lakes and streams throughout acid-sensitive areas of the East is slow (Stoddard et al. 1999; Driscoll et al. 2003; Stoddard et al. 2003). Lakes and streams in the Adirondacks, northern New England, the Appalachian Plateau, the Upper Midwest and western Virginia have been intensively monitored since the early 1980s. A recent analysis indicates that these lakes and streams have shown decreases in concentrations of sulfate, at all sites except western Virginia. This pattern is consistent with decreases in emissions of sulfur dioxide and atmospheric deposition of sulfate. However, these lakes and streams exhibit limited recovery in pH and acid neutralizing capacity, as well as continued acid episodes

(Stoddard et al. 1999; Stoddard et al. 2003). Note, streams in western Virginia show no recovery of acid neutralizing capacity.

Three factors account for the slow chemical recovery of the water quality of acid impacted surface waters, despite the decreased deposition of sulfur associated with the Clean Air Act. First, levels of acid-neutralizing base cations in streams have decreased markedly due to a loss of base cations from the soil and, to a lesser extent, a reduction in atmospheric inputs of base cations. Second, inputs of nitric acid have acidified surface waters and elevated their concentration of nitrate in many regions of the Northeast, particularly the Adirondack and Catskill regions of New York. Finally, sulfur has accumulated in the soil and is now being released to surface water as sulfate, even though sulfate deposition has decreased. It appears the only approach to accelerate the recovery of acid-impacted lakes is to make additional cuts in emissions of sulfur dioxide and nitrogen oxides (Driscoll et al. 2001; Gbondo-Tugbawa and Driscoll 2002; Chen and Driscoll 2005a, b).

The modest decreases in sulfate concentrations and increases in pH and acid neutralizing capacity exhibited in some surface waters is an encouraging sign that impacted ecosystems are responding to emission controls and moving toward chemical recovery. Nevertheless the magnitude of these changes is small compared to the magnitude of increases in sulfate and decreases in acid neutralizing capacity that have occurred in acid-impacted areas following historical increases in acidic deposition.

4.2 Response of aquatic biota to acidification of surface waters by acidic deposition

Decreases in pH and elevated concentrations of dissolved inorganic aluminum have resulted in physiological changes to organisms, direct mortality at sensitive life history stages, and reduced the species diversity and abundance of aquatic life in many streams and lakes in acid-impacted areas of the East. Fish have received the most attention to date, but entire food webs are often adversely affected (Schindler et al. 1985).

Decreases in pH and increases in aluminum concentrations have diminished the species diversity and abundance of plankton, invertebrates, and fish in acid-impacted surface waters in the East (Baker et al. 1990). A detailed summary of the response of aquatic biota to the acidification of surface waters is provided in Table 2.

In the Adirondacks, a significant positive relationship exists between the pH and ANC levels in lakes and the number of fish species present in those lakes (Gallagher and Baker 1990). Surveys of 1,469 Adirondack lakes conducted in 1984 and 1987 show that 24 percent of lakes (i.e., 346) in this region do not support fish (Gallagher and Baker 1990). These lakes had consistently lower pH and ANC, and higher concentrations of aluminum than lakes that contained one or more species of fish. Experimental studies and field observations demonstrate that even acid-tolerant fish species such as brook trout have been eliminated from some waters in New York (Gallagher and Baker 1990).

Similar relationships are evident in streams of the Shenandoah National Park (Bulger et al. 1999). Studies at the Shenandoah National Park demonstrate effects of acidic deposition on fish at three ecosystem levels:

- Effects on single organisms (condition factor- the relationship between the weight and the length of a fish). Studies of blacknose dace indicated that fish condition factor was related to several chemical indicators of acid-base status, including minimum pH. This analysis suggests that fish in acidic streams use more energy to maintain internal chemistry then would otherwise be used for growth (Dennis and Bulger 1999).

- Population -level effects (increased mortality). Bioassay experiments using brook trout eggs and fry showed greater mortality in a chronically acidic stream than in a high acid neutralizing capacity stream (Webb 2004). Eggs and fry are sensitive life history stages for fish.

- Community -level effects (reduced species richness). As observed for the Adirondacks, species richness decreases with decreasing acid neutralizing capacity and pH in streams of the Shenandoah National Park (Bulger et al. 1999).

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

5.0 EFFECTS OF ATMOSPHERIC MERCURY DEPOSITION.

Eastern North America receives elevated atmospheric mercury deposition from a combination of local, regional and global sources. Forested regions with a prevalence of wetlands and unproductive surface waters, combine to promote high concentrations of mercury in freshwater biota, and are particularly sensitive to mercury deposition (Driscoll et al. 2007). Methylmercury that is formed from inorganic mercury in wetlands and sediments, bioaccumulates up the freshwater food chain, typically to be a factor of one-million to ten-million, resulting in exposure to humans and wildlife through fish consumption. Average mercury concentrations in yellow perch fillets exceed the EPA human health criterion across the region. Mercury concentrations are high enough in piscivorous wildlife to cause adverse behavioral, physiological and reproductive effects.

Biological mercury hotspots have been identified in the northeastern U.S. and southeastern Canada using a dataset of biotic Hg concentrations (Evers et al. 2007). Eight layers representing three major taxa and >7,300 observations were used to locate five

biological mercury hotspots and nine areas of concern. The biological mercury hotspots include the western and central Adirondacks, the upper Connecticut River, the Merrimack River, the upper Androscoggin and Kennebec rivers in Maine, and southern and central Nova Scotia. Yellow perch and common loon were chosen as indicator species for human and ecological effects of mercury, respectively. Thresholds of 0.30 μg/g in yellow perch fillets and 3.0 μg/g in common loon blood were used in the analysis. The biological mercury hotspots receive elevated atmospheric mercury deposition, have high landscape sensitivity, and/or experience large reservoir fluctuations. In the Merrimack River watershed local mercury emissions are linked to elevated local deposition and high mercury concentrations in biota. Historic data for this region suggest that mercury emission reductions from local sources can lead to rapid reductions in mercury in biota.

5.1 Linkages between surface water acidification and fish mercury concentrations

Several research studies suggest a linkage between acidic deposition and mercury levels in fish. Atmospheric deposition of sulfate associated with sulfur dioxide emissions provides the necessary substrate for methylating bacteria (Gilmour et al. 1992). Methylating bacteria convert inorganic mercury into methyl mercury, the form of mercury which bioaccumulates in fish.

The role of sulfate in the production of methyl mercury is under investigation by a team of researchers in Minnesota. Jeremiason et al. (2006) experimentally added sulfate to a wetland, observing increased methylation and increased export of methyl mercury. They inferred that increasing sulfur dioxide emissions and sulfate deposition would result in increases in methyl mercury in the fish of receiving waters (Jeremiason et al. 2006). Similar experiments have been conducted in Sweden and Canada (Branfireun et al. 1999, 2001).

Studies across eastern North America have shown that many lakes and reservoirs have elevated concentrations of mercury in fish tissue (Grieb et al. 1990; Suns and Hitchin 1990; Driscoll et al. 1994; Kamman et al. 2004, 2005; Driscoll et al. 2007). Many of these surface waters have fish mercury content above the U.S. Environmental Protection Agency health advisory action level of 0.3 μg/g. In addition numerous studies across eastern North America have also reported increases in fish mercury concentrations with decreases in surface water pH. (Grieb et al. 1990; Suns and Hitchin 1990; Driscoll et al. 1994; Kamman et al. 2004; Kamman et al. 2005; Driscoll et al. 2007).

Hrabik and Watras (2002) used reference data and data from an experimentally acidified lake to examine the relative contribution of atmospheric mercury deposition and acidic deposition in regulating changes in fish mercury concentrations. They observed that decreases in fish mercury in an experimentally de-acidified basin exceeded those in the reference basin. Specifically, they found that approximately one-half of the change in fish mercury concentrations over a six-year period could be attributed to de-acidification (Hrabik and Watras 2002). This study suggests that acidification of lakes by acidic

deposition has enhanced fish mercury concentrations and that concentrations of mercury in fish are likely to decrease with decreasing acidic deposition.

6.0 Monitoring Acidic and Mercury Deposition and the Ecological Effects of These Contaminants on Surface Waters

6.1 Monitoring effects of acidic deposition

Environmental monitoring is a critical tool to help track the effectiveness of past controls on emissions of air pollutants and to guide future air quality management in the U.S. (NAS 2004). There are several national programs which are widely used to evaluate the extent and change in atmospheric deposition and to assess changes in surface water chemistry in response to changes in acidic deposition. In the U.S. wet deposition has been monitored at more than 200 sites, by both independent researchers and the inter-agency National Atmospheric Deposition Program/National Trends Network (http://nadp.sws.uiuc.edu/). Dry deposition is monitored at 97 sites in the U.S. by the U.S. Environmental Protection Agency (EPA) Clean Air Status and Trends Network (http://www.epa.gov/castnet/.

The US EPA currently also funds two surface water programs as part of the Environmental Monitoring and Assessment Program: (1) Long-Term Monitoring Program (LTM), and (2) Temporally Integrated Monitoring of Ecosystems (TIME). Together these two monitoring programs provide important information on changes in water chemistry in response to the deposition of air pollutants. These programs fund the sampling of surface water quality in the eastern U.S in part to determine whether and to what extent sensitive ecosystems are responding to changes in the acidity of precipitation as a result of the Acid Rain Program within Title IV of the Clean Air Act Amendments of 1990. This combined program (sometimes referred to as LTM/TIME) is the most geographically extensive network of sites in the U.S. that is tracking whether sensitive ecosystems are recovering from the decreasing the acidity of precipitation since the early 1980s.

Through collection of water chemistry data at regular intervals and in sensitive ecosystems in several regions that includes New England, the Adirondacks, the Appalachian Plateau, and western Virginia, the value of this program has repeatedly been demonstrated through high-profile papers and reports published in the scientific literature as well as in scientific reports that have informed the policy community. For example, several papers published in the peer-reviewed literature in recent years have used LTM/TIME data to examine the issue of recovery in sensitive ecosystems (Driscoll et al., 2003; Kahl et al., 2004; Webb et al., 2004). Recent policy-informing reports that have used LTM/TIME data include an assessment of the response of surface-water chemistry to the 1990 Clean Air Act Amendments (Stoddard et al., 2003) and the most recent report of the federal inter-agency National Acid Precipitation Assessment Program to the U.S. Congress (NAPAP, 2005).

The Clean Air Interstate Rule (CAIR) is currently being implemented by the EPA in part to bring about additional decreases in emissions of sulfur dioxide and nitrogen oxides and to foster the recovery of previously acidified ecosystems. This rule will result in additional emissions reductions beginning in 2010 and continuing through 2015. Data from LTM/TIME program collected during this period when combined with previous data will provide critical information on the effects of these emissions cuts in sensitive ecosystems.

Unfortunately, there are two items in the President's 2008 budget that pose a challenge to those of us in the acid rain research and policy communities. This budget shows a $1 million cut to the CASTNet program and a cut of $5.75 million (effectively zeroing out the program) to the extramural EMAP program---this item supports both the LTM and TIME programs of EPA. Without these two critical monitoring programs it will be difficult if not impossible to track the response atmospheric chemistry and acid-sensitive surface waters current and future controls on emissions of air pollutants.

6.2 Monitoring effects of mercury deposition

There is also a critical need to be able to assess the impacts of past and future controls on emissions of mercury in the U.S. Unlike acidic deposition the monitoring infrastructure has not been developed to monitor ecosystem effects of mercury deposition. In the U.S. and Canada, measurements of wet mercury deposition are largely made through the Mercury Deposition Network (MDN; http://nadp.sws.uiuc.edu/mdn/). The MDN shows that wet mercury deposition is highest in the Southeast (i.e., Florida, Mississippi) and lowest in the West.

While the data and information discussed this testimony provide insights into the nature and extent of mercury pollution across the Northeast, many data gaps remain and most other regions of the United States and Canada do not have such a large biological data set from which to evaluate biological mercury hotspots. In addition, the current Mercury Deposition Network is too sparse and limited by its focus on wet deposition in rural areas to provide a detailed understanding of deposition patterns and their connection to local sources in the United States. In sensitive forest ecosystems most of the mercury inputs are derived from (i.e., as particles or gases), so a wet deposition network is not completely effective in tracking total mercury loading.

The problem of insufficient mercury monitoring was recently echoed in a report by the EPA Inspector General which states, "Without field data from an improved monitoring network...'utility-attributable' hotspots that pose health risks may occur and go undetected." The reports goes on to say: "We recommend that EPA develop and implement a mercury monitoring plan to (1) assess the impact of Clean Air Mercury Rule (CAMR) on mercury deposition and fish tissue; and (2) evaluate and refine mercury estimation tools and models" (EPA 2006).

A comprehensive long-term mercury monitoring program focused on mercury deposition, watershed cycling, and biological effects would allow scientists to conduct a national scale assessment of biological mercury hotspots, and to link changes in emissions and deposition with ecosystem effects and response. At present, scientists must rely on limited information to make these important linkages. Increased mercury monitoring should extend to forest ecosystems where recent research revealed elevated mercury in insect-eating songbirds (Evers et al. 2005, Rimmer et al. 2005) but little information exists to determine how mercury accumulates in terrestrial food webs and what levels are harmful to these birds.

A roadmap for a comprehensive national mercury monitoring program was developed by a team of scientists and is detailed Mason et al. (2005). The proposed program emerged from an EPA workshop in 2003 that brought together 32 scientists from across the United States and a number of other countries to devise a national mercury monitoring program and was recently published in Harris et al. (2007). The critical need for a comprehensive ecosystem mercury monitoring program was recently emphasized by bills introduced in both the House of Representatives and the Senate to establish such a program through a multi-agency initiative (House - H.R. 1533 "Comprehensive National Mercury Monitoring Establishment Act" Sponor: Tom Allen (D-ME). Cosponsors: James Walsh (R-NY), John McHugh (NY), Michael Michaud (ME), Mark Steven Kirk (IL), Raul Grijavla (AZ); Senate - S. 843 "Comprehensive National Mercury Monitoring Act" Sponsor: Susan Collins (R-ME), Joseph Lieberman (R-CT), Hillary Clinton (D-NY)).

Table 2. Biological effects of surface water acidification (after Baker et al. 1990).

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

REFERENCES

Bailey SW, Hornbeck JM, Driscoll CT, Gaudette HE. 1996. Calcium inputs and transport in a base-poor forest ecosystem as interpreted by Sr isotopes. Water Resources Research 32:707-719.

Baker JP, Gherini SA, Christensen SW, Driscoll CT, Gallagher J, Munson RK, Newton RM. 1990. Adirondack Lake Survey: An interpretive analysis of fish communities and water chemistry, 1984-87. Adirondack Lakes Survey Corporation, Ray Brook, NY.

Baker JP, Van Sickle J, Gagen CJ, DeWalle Jr DR, DeWalle DR, Sharpe WF, Carline RF, Baldigo BP, Murdoch PS, Bath DW, Kretser WA, Simonin HA, Wigington PJ. 1996. Episodic acidification of small streams in the northeastern United States: effects on fish populations. Ecological Applications 6:422-437.

Brakke DF, Henriksen A, Norton SA. 1989. Estimated background concentrations of sulfate in dilute lakes. Water Resource Bulletin 25:247.

Branfireun BA, Roulet NT, Kelly CA, Rudd JWM. 1999. In situ sulphate stimulation of mercury methylation in a boreal peatland: Toward a link between acid rain and methylmercury contamination in remote environments (Paper 1999GB900033). Global Biogeochemical Cycles 13:8.

Branfireun BA, Bishop K, Roulet NT, Granberg G, Nilsson M. 2001. Hydrology and Biogeochemical Cycles - Mercury cycling in boreal ecosystems: The long-term effect of acid rain constituents on peatland pore water methylmercury concentrations (Paper 2000GL011867). Geophysical Research Letters 28:4.

Bulger AB, Cosby BJ, Dolloff CA, Eshleman KN, Galloway JN, Webb JR. 1999. Shenandoah National Park Fish in Sensitive Habitats (SNP FISH) Project Final Report: Volume I-IV plus interactive computer model. Submitted to National Park Service, Luray, VA. (Available on CD-Rom from John F. Karish, National Park Service, Northeast Field Area, 209B Ferguson Building, University Park, PA 16802-4301, U.S.A.)

Butler TJ, Likens GE, Stunder BJ. 2001. Regional-scale impacts of Phase I of the Clean Air Act Amendments: the relationship between emissions and concentrations, both wet and dry. Atmospheric Environment 35:1015-1028.

Butler TJ, Likens GE, Vermeylen FM, Stunder BJB. 2003. The relation between NO_x emissions and precipitation NO_3^- in the eastern USA. Atmospheric Environment 37:2093-2104.

Castro MS, and Driscoll CT. 2002. Atmospheric nitrogen deposition to estuaries in the mid-Atlantic and northeastern United States. Environmental Science and Technology 36:3242-3249.

Charles DF, ed. 1991. Acidic deposition and aquatic ecosystems. regional case studies. New York: Springer-Verlag New York, Inc.

Chen L, Driscoll CT. 2005. Regional application of an integrated biogeochemical model to northern New England and Maine. Ecological Applications 15:1783-1797.

Chen L, Driscoll CT. 2005. Regional assessment of the response of the acid-base status of lake watersheds in the Adirondack region of New York to changes in atmospheric deposition using PnET-BGC. Environmental Science and Technology 39:787-794.

Cronan, CS, Schofield, CL. 1990. Relationships between aqueous aluminum and acidic deposition in forested watersheds of North America and Northern Europe. Environmental Science and Technology 24:1100-1105.

Cronan CS, Grigal DF. 1995. Use of calcium/aluminum ratios as indicators of stress in forest ecosystems. Journal of Environmental Quality 24:209-226.

Dennis TE., Bulger AJ. 1999. The susceptibility of blacknose dace (*Rhinichthys atratulus*) to acidification in Shenandoah National Park. In: A.B. Bulger, B.J. Cosby, C.A. Dolloff, K.N. Eshleman, J.N. Galloway, and J.R. Webb. Shenandoah National Park Fish in Sensitive Habitats. Project Final Report, Volume IV, Chapter 6B.

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

Driscoll CT, Newton RM, Gubala CP, Baker JP, Christensen S. 1991. Adirondack Mountains. In: D.F. Charles (ed.) Acidic Deposition and Aquatic Ecosystems: Regional Case Studies. Springer-Verlag, New York. pp. 133-202.

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

Driscoll CT, Postek KM. 1995. The chemistry of aluminum in surface waters. In: G. Sposito (ed.) The Environmental Chemistry of Aluminum, Lewis Publishers, Chelsea, MI, pp. 363-418.

Driscoll CT, Lawrence GB, Bulger AJ, Butler TJ, Cronan CS, Eagar C, Lambert KF, Likens GE, Stoddard JL, Weathers KC. 2001. Acidic deposition in the northeastern U.S.: sources and inputs, ecosystems effects, and management strategies. BioScience 51:180-198.

Driscoll CT, Driscoll KM, Mitchell MJ, Raynal DJ. 2003a. Effects of acidic deposition on forest and aquatic ecosystems in New York State. Environmental Pollution 123:327-336.

Driscoll CT, Driscoll KM, Roy KM, Mitchell, MJ. 2003d. Chemical response of lakes in the Adirondack Region of New York to declines in acidic deposition. Environmental Science and Technology 37:2036-2042.

Driscoll CT, Han Y-J, Chen CY, Evers DC, Lambert KF, Holsen TM, Kamman NC, Munson RK. 2007. Mercury contamination in forest and aquatic ecosystems in the Northeastern U.S.: Sources, transformations and management options. BioScience 57:17-28.

Evers, D.C., N.M.Burgess, L. Champoux, B. Hoskins, A. Major, W.M. Goodale, R.J. Taylor, R.H. Poggenga and T. Daigle. 2005. Patterns and interpretation of mercury exposure in freshwater avian communities in northeastern North America. Ecotoxicology. 14: 193-222.

Evers, D.C., Y-J. Han, C.T. Driscoll, N.C. Kamman, W.M. Goodale, K.F. Lambert, T.M. Holsen, C.Y. Chen, T.A. Clair and T. Butler. 2007. Biological mercury hotspots in the Northeastern United States and Southeastern Canada. BioScience 57:1-15.

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

Gbondo-Tugbawa SS, Driscoll CT. 2002. Evaluation of the effects of future controls on sulfur dioxide and nitrogen oxide emissions on the acid-base status of a northern forest ecosystem. Atmospheric Environment 36:1631-1643.

Gilmour CC, Henry EA, Mitchell R. 1992. Sulfate stimulation of mercury methylation in freshwater sediments. Environmental Science and Technology 26:2281-2287.

Gorham E. 1989. Scientific understanding of ecosystem acidification: a historical review. Ambio 3:150-154.

Grieb, TM., Driscoll CT, Gloss SP, Schofield CL, Bowie GL, Porcella DB. 1990. Factors affecting mercury accumulation in fish in the Upper Michigan Peninsula. Environmental Toxicology and Chemistry. 9:919-930.

Gunthardt-Goerg MS, McQuattie CJ, Mauer S, Frey B. 2000. Visible and microscopic injury in leaves of five deciduous tree species related to current critical ozone levels. Environmental Pollution 109: 489-500.

Harris, R., D.P. Krabbenhoft, R. Mason, M.W. Murray, R. Reash and T. Saltman (eds.) 2007Ecosystem Responses to Mercury Comtamination: Indicators of Change, Taylor and Francis, Boco Rotan, FL., 216 p.

Hrabik TR, Watras CJ. 2002. Recent declines in mercury concentration in a freshwater fishery: Isolating the effects of de-acidification and decreased atmospheric mercury deposition in Little Rock Lake. The Science of the Total Environment 297:229-237.

Jaworski NA, Howarth RW, Hetling LJ. 1997. Atmospheric deposition of nitrogen oxides onto the landscape contributes to coastal eutrophication in the Northeast United States. Environmental Science and Technology 31:1995-2005.

Jeremiason J.D., D.R. Engstrom, E.B. Swain, E.A. Nater, B.M. Johnson, J.E. Almendinger, B.A. Monson, Kolka, R.K. 2006. Sulfate addition increases methylmercury production in an experimental wetland. Environmental Science and Technology 40:3800-3806.

Kahl JS, and 15 others. 2004. Have U.S. surface waters responded to the 1990 Clean Air Act Amendments? Environmental Science and Technology 38: 484A – 490A.

Kamman NC, Lorey PM, Driscoll CT, Estabrook R, Major A, Pientka B, Glassford E. 2004. Assessment of mercury in waters, sediments, and biota of New Hampshire and Vermont lakes, USA, sampled using a geographically randomized design. Environmental Toxicology and Chemistry 23:1172-1186.

Kamman NC, Burgess NM, Driscoll CT, Simonin HA, Goodale W, Linehan J, Estabrook R, Hutcheson M, Major A, Scheuhammer AM, Scruton DA. 2005. Mercury in freshwater fish of northeast North America - a geographic perspective based on fish tissue monitoring databases. Ecotoxicology 14:163-180.

Kretser W, Gallagher J, Nicolette J. 1989. Adirondack Lakes Study. 1984-1987. An evaluation of fish communities and water chemistry. Ray Brook (NY): Adirondacks Lakes Survey Corporation.

Lawrence GB, David MB, Lovett GM, Murdoch PS, Burns PA, Baldigo BP, Thompson AW, Porter JH, Stoddard JL. 1999. Soil calcium status and the response of stream chemistry to changing acidic deposition rates in the Catskill Mountains of New York. Ecological Applications 9:1059-1072.

Likens GE, Driscoll CT, Buso DC. 1996. Long-term effects of acid rain: response and recovery of a forested ecosystem. Science 272:244-246.

Likens GE, Driscoll CT, Buso DC, Siccama TG, Johnson CE, Lovett GM, Fahey TJ, Reiners WA, Ryan DF, Martin CW, Bailey SW. 1998. The biogeochemistry of calcium at Hubbard. Biogeochemistry 41:89-173.

Likens GE, Butler TJ, Buso DC. 2000. Long- and short-term changes in sulfate deposition: effects of the 1990 Clean Air Act Amendments. Biogeochemistry 52:1-11.

Lynch JA, Bowersox VC, Grimm JW. 2000. Changes in sulfate deposition in eastern USA following implementation of Phase I of Title IV of the Clean Air Act Amendments of 1990. Atmospheric Environment 34:1665-1680.

MacAvoy SE, Bulger AJ. 1995. Survival of brook trout (*Salvelinus fontinalis*) embryos and fry in streams of different acid sensitivity in Shenandoah National Park, USA. Water Air and Soil Pollution 85: 439-444.

Malm WC, Sisler JF, Huffman D, Eldred RA, Cahill TA. 1994. Spatial and seasonal trends in particle concentration and optical extinction in the United States. Journal of Geophysical Research 99:1347-1370.

Mason, R.P., M.L. Abbott, R.A. Bodaly, O.R. Bullock, Jr., C.T. Driscoll, D. Evers, S.E. Lindberg, M. Murray, and E.B. Swain. 2005. Monitoring the response to changing mercury deposition. Environ. Sci. Technol. 39:15-22A.

[NAPAP] National Acid Precipitation Assessment Program. 1998. NAPAP Biennial Report to Congress. An Integrated Assessment. National Acid Precipitation Program. Washington (DC).

[NAPAP] National Acid Precipitation Assessment Program. 2005. National Acid Precipitation Assessment Program Report to Congress: An Integrated Assessment, Washington, DC, 85 p.

[NRC] National Research Council 2004. Air Quality Management in the United States, NRC Press, Washington D.C., 401 p.

Paerl, HW, Boynton WR, Dennis RL, Driscoll CT, Greening HS, Kremer JN, Rabalais NN, and Seitzinger SP. 2000. Atmospheric deposition of nitrogen in coastal waters: biogeochemical and ecological implications. In: R. Valigura, et al. (eds.) Nitrogen Loading in Coastal Water Bodies: An Atmospheric Perspective. American Geophysical Union, Washington, DC, pp. 11-52.

Reuss JO, Johnson DW. 1986. Acid Deposition and the Acidification of Soils and Waters. Springer-Verlag, New York.

Rimmer, C.C., K.P. McFarland, D.C. Evers, E.K. Miller, Y. Aubry, D. Bushey, and R.J. Taylor. Mercury concentrations in Bicknell's thrush and other insectivorous passerines in montane forests of northeastern North America. Ecotoxicology 14: 223-240.

Schindler DW, Mills KH, Malley DF, Findlay S, Shearer JA, Davies IJ, Turner MA, Lindsey GA, Cruikshank DR. 1985. Long-term ecosystem stress: Effects of years of

experimental acidification. Canadian Journal of Fisheries and Aquatic Science 37:342-354.

Stoddard JL, Jeffries DS, Lukewille A, Clair TA, Dillon PJ, et al. 1999. Regional trends in aquatic recovery from acidification in North America and Europe. Nature 401:575-578.

Stoddard JL, Kahl JS, Deviney FA, DeWalle DR, Driscoll CT, Herlihy AT, Kellogg JH, Murdoch PS, Webb JR, Webster KE. 2003. Response of surface water chemistry to the Clean Air Act Amendments of 1990. Washington, D.C.

Suns K., Hitchin G. 1990. Interrelationships between mercury levels in yearling yellow perch, fish condition and water quality. Water, Air, and Soil Pollution 50:255-265.

Van Sickle J, Baker JP, Simonin HA, Baldigo BP, Kretser WA, Sharpe WF. 1996. Episodic acidification of small streams in the northeastern United States: fish mortality in field bioassays. Ecological Applications 6:408-421.

Warby, R.A.F., C.E. Johnson, and C.T. Driscoll. 2005. Chemical recovery of surface waters across the northeastern U.S. from reduced inputs of acidic deposition: 1984-2001. Environ. Sci. Technol. 39:6548-6554.

Warby, R.A.F., C.E. Johnson, and C.T. Driscoll. Organic soils across the northeastern U.S.A. are continuing to acidify, despite reductions in acidic deposition across the region: 1984 – 2001. J. Soil. Sci. Am. (in review).

Webb R. 2004. Effects of acidic deposition on aquatic resources in the Central Appalachian Mountains. A Shenandoah Watershed Study Report. http://swas.evsc.virginia.edu .

Webb, JR, Cosby, BJ, Deviney, FA, Galloway, JN, Maben, SW, Bulger, AJ. 2004. Are brook trout streams in western Virginia and Shenandoah Park recovering from acidification? Environmental Science and Technology **38**: 4091 – 4096.

Weiner JG, Krabbenhoft DP, Heinz GH, Scheuhammer AM. 2003. Ecotoxicology of mercury. In: Handbook of Ecotoxicology. Hoffman DJ et al. (eds), Lewis Publishers, Boca Raton, FL pp. 409-462.

Wigington Jr PJ, Baker JP, DeWalle DR, Kretser WA, Murdoch PS, Simonin HA, Van Sickle J, McDowell MK, Peck DV, Barchet WR. 1996. Episodic acidification of small streams in the northeastern United States: Episodic Response Project. Ecological Applications 6:374-388.