



Air Quality Related Values (AQRVs) for Northeast Temperate Network (NETN) Parks

Effects from Ozone; Visibility Reducing Particles; and Atmospheric Deposition of Acids, Nutrients and Toxics

Natural Resource Report NPS/NETN/NRR—2016/1166



ON THE COVER

Photograph of air quality related values within various National Park units. Wildflowers, clear views, aquatic species, and lichens may all be threatened by air pollution.

Photographs courtesy of the National Park Service

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Summary

This report describes the Air Quality Related Values (AQRVs) of the Northeast Temperate Network (NETN). AQRVs are those resources sensitive to air quality and include streams, lakes, soils, vegetation, fish and wildlife, and visibility. The NETN parks that are included in the NPS Inventory and Monitoring (I&M) Program, and discussed in this report, are Acadia National Park (ACAD), Boston Harbor Islands National Recreation Area (BOHA), Marsh-Billings-Rockefeller National Historical Park (MABI), Minute Man National Historical Park (MIMA), Morristown National Historical Park (MORR), Roosevelt-Vanderbilt national historic sites (ROVA; consisting of Eleanor Roosevelt National Historic Site [ELRO], Home of Franklin D. Roosevelt National Historic Site [HOFR], and Vanderbilt Mansion National Historic Site [VAMA]), Saint-Gaudens National Historic Site (SAGA), Saugus Iron Works National Historic Site (SAIR), and Weir Farm National Historic Site (WEFA). The network region also includes parts of the Appalachian National Scenic Trail, which runs from Maine to Georgia; the trail corridor is addressed in the companion report for the Appalachian Highlands Network. Only limited data are available for some of the small parks in the NETN, including ELRO, which is not discussed in this report. ACAD is designated as Class I, giving it a heightened level of protection against harm caused by poor air quality under the Clean Air Act (CAA).

Sullivan et al. (2011a, 2011b) and Kohut (2007) conducted risk assessments for acidification, eutrophication, and ozone (O_3) for the NCCN parks; their results are described in this report. This report also describes air pollutant emissions and air quality, and their effects on AQRVs. The primary pollutants likely to affect AQRVs include nitrogen (N) and sulfur (S) compounds (nitrate [NO_3^-], ammonium [NH_4^+], and sulfate [SO_4^{2-}]); ground-level O_3 ; haze-causing particles; and airborne toxics. Background for this section can be found in “Air Quality Related Values (AQRVs) in National Parks: Effects from Ozone; Visibility Reducing Particles; and Atmospheric Deposition of Acids, Nutrients and Toxics” (Sullivan 2016).

Parks in the NETN include several near heavily urbanized areas around New York City and Boston, as well as parks in relatively undeveloped areas in northern New York, New Hampshire, Vermont, and Maine. Air pollution may be an important stressor to natural resources in the NETN parks. Sulfur and N pollutants can cause acidification of streams, lakes, and soils. Nitrogen pollutants can also cause undesirable enrichment of natural ecosystems, leading to changes in plant species diversity and soil nutrient cycling. Portions of the network region away from the coastal corridor, and also the northeastern corner of the network, generally have relatively low S emissions, less than 1 ton per square mile per year ($ton/mi^2/yr$) in most counties. Emissions of S into the atmosphere in New York and states that lie up-wind from the extensively studied Adirondack and Catskill mountain regions of New York were very high in past decades, but have been gradually decreasing since the 1970s. County-level N emissions within the network region ranged from less than 1 $ton/mi^2/yr$, mainly in the north, to greater than 100 $tons/mi^2/yr$ in some of the densely urbanized areas in and around Boston and near New York City. Total S and total N deposition within the NETN in 2002 were also both highly variable from north to south.

Atmospheric deposition of both S and N were higher than 10 kilograms per hectare per year (kg/ha/yr) at many of the NETN parks in 2001. Deposition of S and oxidized N have decreased substantially at NETN parks since that time. Deposition of reduced N decreased between 2001 and 2011 at some parks in NETN and increased at others.

Substantial research on air pollution effects has been conducted in ACAD, but not in other parks within the network. ACAD's steep slopes, thin soils, high mountains, and exposure to coastal fog increase the park's susceptibility to deposition of long-range atmospheric pollutants, including S, N, mercury (Hg), and other contaminants. Most of ACAD is underlain by granite. The relatively low buffering capacity of the overlying soils, together with areas having steep slopes, and in some areas also relatively high elevation, influence susceptibility of soil and drainage water to acidification. Portions of the Adirondack and Catskill mountains and the New England uplands are among the areas of the United States most affected by acidification from acidic deposition. Several NETN parks (i.e., SARA, MABI, and SAGA) are located close to this highly impacted portion of the NETN region.

The health of sugar maple (*Acer saccharum*) trees is strongly influenced by the acid-base chemistry, especially the availability of calcium (Ca), in soil. Trees that grow on soils having low base cation supply are stressed and consequently can become more susceptible to damage from defoliating insects, drought, and extreme weather. Sugar maples occur in all of the NETN parks.

Although upwind acidifying emissions have been reduced since passage of the 1990 CAA, there has been little corresponding change in the acid neutralizing capacity (ANC) and pH of surface waters in ACAD. Some episodic acidification of headwater streams likely occurs in ACAD as a result of S, N, and marine salt and fog deposition to thin, acidic soils. Salt inputs to surface waters at ACAD can occur due to marine aerosol deposition and/or road salt application.

Nutrient enrichment in response to atmospheric N deposition is an important research and management issue at some locations in the Northeast. In some areas which have moved towards a condition of N saturation, high levels of N deposition have contributed to elevated concentrations of NO_3^- in drainage waters. Elevated NO_3^- leaching causes depletion of base cations from forest soils, with adverse effects on sensitive tree species and acidification of drainage waters in base-poor soils. Forests considered especially sensitive to N deposition include high-elevation red spruce (*Picea rubens*) and sugar maple forests. Effects of N deposition on root allocation or late-season growth may exacerbate other stresses from acidic deposition and harsh climate. Base cation depletion of soil may be associated with an increased likelihood of Al toxicity to plants and the decline of red spruce and/or sugar maple trees.

Several streams in ACAD have been shown to have chronically elevated NO_3^- concentrations (Johnson et al. 2007, Nelson et al. 2008), which suggests possible N-saturation of ACAD forests (Kahl et al. 2007a). However, Miller et al. (2014) suggested that soils in ACAD were not very sensitive to N saturation, based on having high soil organic matter. Other parks in the NETN have lower soil organic matter and may be more sensitive to N saturation in response to N deposition (Miller et al. 2014).

Ozone pollution can harm human health, reduce plant growth, and cause visible injury to foliage. Two of the NETN parks, MORR and WEFA, are in counties designated by EPA as nonattainment for the O₃ standard because of their high O₃ levels. The highest O₃ concentrations in the United States in 2007 were located in six states, including Massachusetts, which contains several NETN parks. Urban-derived plumes of O₃ have been tracked from the Boston and New York City areas to remote parks like ACAD. Ozone impacts to vegetation have been studied in ACAD, which has experienced periodic episodes of elevated ground level O₃ throughout the summer months. Foliar symptoms of O₃ injury were observed in several plant species in ACAD. Ozone injury to plants has not been evaluated in the other NETN parks.

Particulate pollution can cause haze, reducing visibility. Levels of haze are high at times in the NETN parks. Sulfate is the most important cause of fine particle pollution and visibility impairment across the Mid-Atlantic/Northeast Visibility Union (MANE-VU) states, including states with NETN parks. At the Class I sites (including ACAD and several Forest Service and U.S. Fish and Wildlife Service wilderness areas) in this region, SO₄²⁻ accounts for about one-half to two-thirds of atmospheric total fine particle mass on the 20% haziest days. Although MANE-VU is focusing primarily on fine particle SO₄²⁻ abatement in its initial efforts toward compliance with the Regional Haze Rule (RHR), controls on other haze-producing constituents, such as organic carbon (C) and both urban and mobile sources of nitrogen oxides (NO_x) during winter will also be important.

Haze is monitored at ACAD; in addition, data from the Cape Cod National Seashore (CACO) Interagency Monitoring of Protected Visual Environments (IMPROVE) Network monitor, which is in the adjacent Northeast Coastal and Barrier Network, is considered by EPA to be representative of visibility conditions at BOHA and SAIR. Levels of ambient haze in ACAD, BOHA, and SAIR as represented by measurements at the IMPROVE sites were High for all groups (clearest, haziest, average) and were about 10 deciviews (dv) higher than natural haze on the 20% haziest days.

Airborne contaminants, including Hg, can accumulate in food webs, reaching toxic levels in top predators. Substantial research has been conducted in ACAD on Hg methylation, the influence of SO₄²⁻ on methylation rates, and controls on Hg transport within watersheds. Ecosystems in ACAD are especially sensitive to Hg bioaccumulation, due in part to relatively high Hg deposition and in particular due to watershed and lake characteristics that enhance Hg transport, methylation, and bioaccumulation.

In the northeastern United States, including in and near NETN parks, high concentrations of Hg in yellow perch (*Perca flavescens*) and common loon (*Gavia immer*) have been shown to be significantly correlated with aspects of water chemistry, including total phosphorus (P), dissolved organic carbon (DOC), and ANC. High fish Hg concentrations have also been shown to be correlated with several landscape characteristics: wetland abundance, low lake:watershed area ratio, high percent forest cover, and high atmospheric Hg deposition. These characteristics affect Hg transport, methylation, and trophic transfer. Methylation is critical to the effects of Hg on aquatic biota and piscivorous wildlife; methylmercury (MeHg) is toxic, bioavailable, and accumulates in top predators to levels of concern for both human health and the environment.

Accumulation of MeHg in fish-eating birds can result in damage to nervous, excretory, and reproductive systems. Reduced clutch size, increased number of eggs laid outside the nest, eggshell thinning, and increased embryo mortality have all been documented. Wetland and associated watershed food webs in ACAD are highly susceptible to bioaccumulation of MeHg. Effects of toxic Hg and Al are thought to be causes of the dramatic and perhaps irreversible decline in ACAD of the northern dusky salamander (*Desmognathus fuscus fuscus*), whose diet includes Hg-contaminated prey.

Background

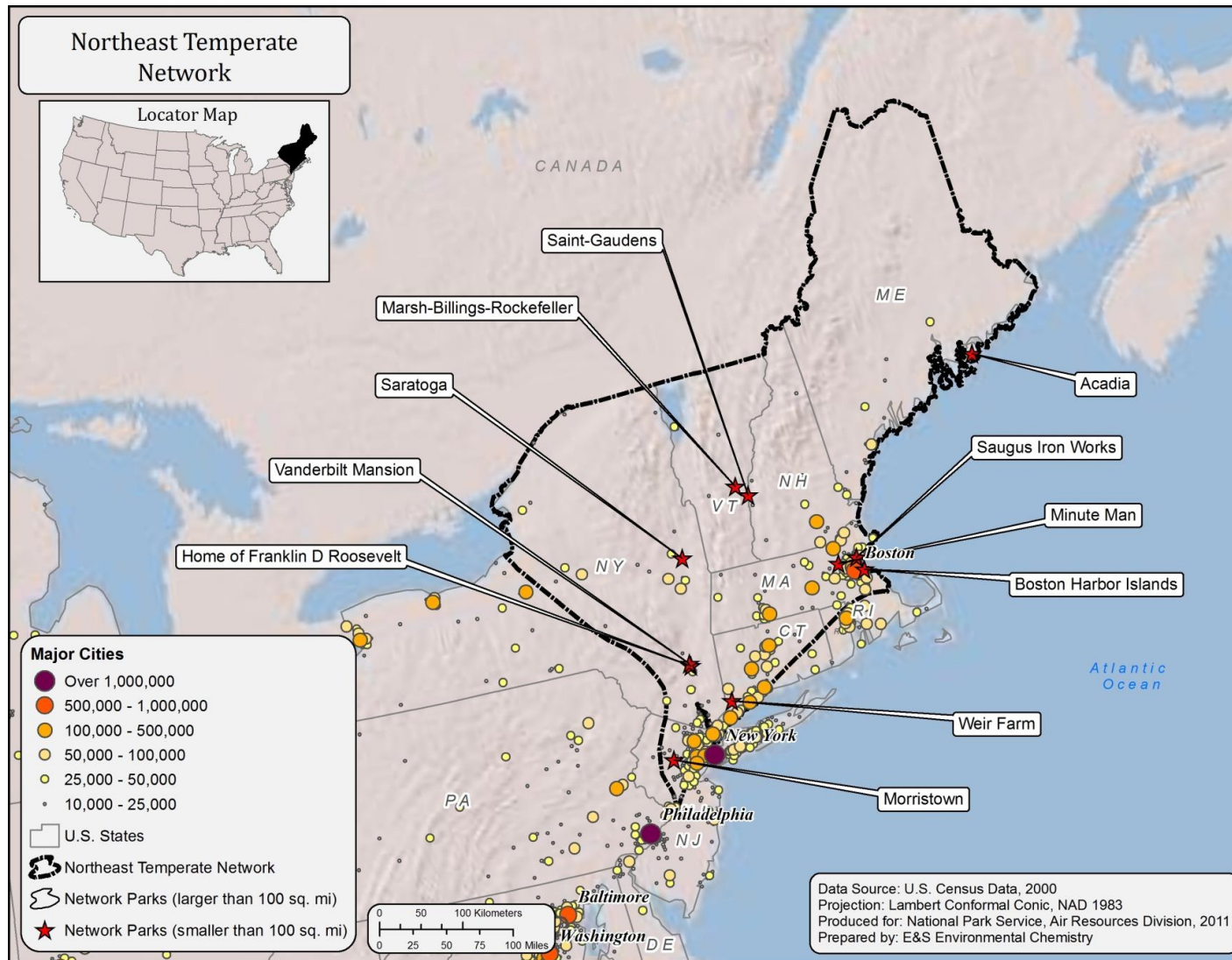
There are 13 Inventory and Monitoring (I&M) parks in the Northeast Temperate Network (NETN), 11 of which are addressed in this report (all except the Appalachian National Scenic Trail [APPA] and Eleanor Roosevelt National Historic Site [ELRO]): Acadia National Park (ACAD), Boston Harbor Islands National Recreation Area (BOHA), Marsh-Billings-Rockefeller National Historical Park (MABI), Minute Man National Historical Park (MIMA), Morristown National Historical Park (MORR), Roosevelt-Vanderbilt national historic sites (ROVA; consisting of ELRO, Home of Franklin D. Roosevelt National Historic Site [HOFR], and Vanderbilt Mansion National Historic Site [VAMA]), Saint-Gaudens National Historic Site (SAGA), Saugus Iron Works National Historic Site (SAIR), and Weir Farm National Historic Site (WEFA). The APPA extends from Maine to Georgia and is addressed in the companion report for the Appalachian Highlands Network.

The largest park in the NETN is ACAD, which is the oldest established national park in the eastern United States. None of the parks in this network are larger than 100 square miles. Some parks in the network are near heavily urbanized areas around New York City and Boston, whereas others are located in relatively undeveloped areas in northern New York, New Hampshire, Vermont, and Maine. There are many medium to large urban population centers in the Northeast, especially along the coastal corridor between Boston and Washington, DC. Elsewhere in the network region, there are fewer population centers larger than 50,000 people. Map 1 shows the network boundary, the location of each park, and population centers having more than 10,000 people.

Atmospheric Emissions and Deposition

Annual county-level sulfur (S) emissions in the NETN region in 2002 were mostly less than 20 tons per square mile ($\text{tons}/\text{mi}^2/\text{yr}$), with only a few small areas having higher levels (Sullivan et al. 2011b). Portions of the network region away from the coastal corridor, and the northeastern corner of the network region, generally had low S emissions, less than $1 \text{ ton}/\text{mi}^2/\text{yr}$ in most counties. There were many point sources of sulfur dioxide (SO_2) within the NETN region, predominantly in the heavily urbanized areas. Most emitted less than 5,000 tons of S per year. Emissions of S into the atmosphere in New York and states that lie up-wind from the Adirondack and Catskill mountain regions were very high in past decades, but have been gradually decreasing since the 1970s.

County-level nitrogen (N) emissions within the network region ranged from less than $1 \text{ ton}/\text{mi}^2/\text{yr}$, mainly in the north, to greater than $100 \text{ tons}/\text{mi}^2/\text{yr}$ in some of the densely urbanized areas in and around Boston and near New York City. However, annual county N emissions were generally less



Map 1. Network boundary and locations of parks and population centers greater than 10,000 people within the NETN region.

than 20 tons/mi²/yr throughout all but the most heavily urbanized areas. There were few N point sources larger than 500 to 1,000 tons per year within this network region. Emissions of N were at relatively constant and high levels during the 1980s and 1990s, but have begun to decline since about 2000 (Sullivan et al. 2011b).

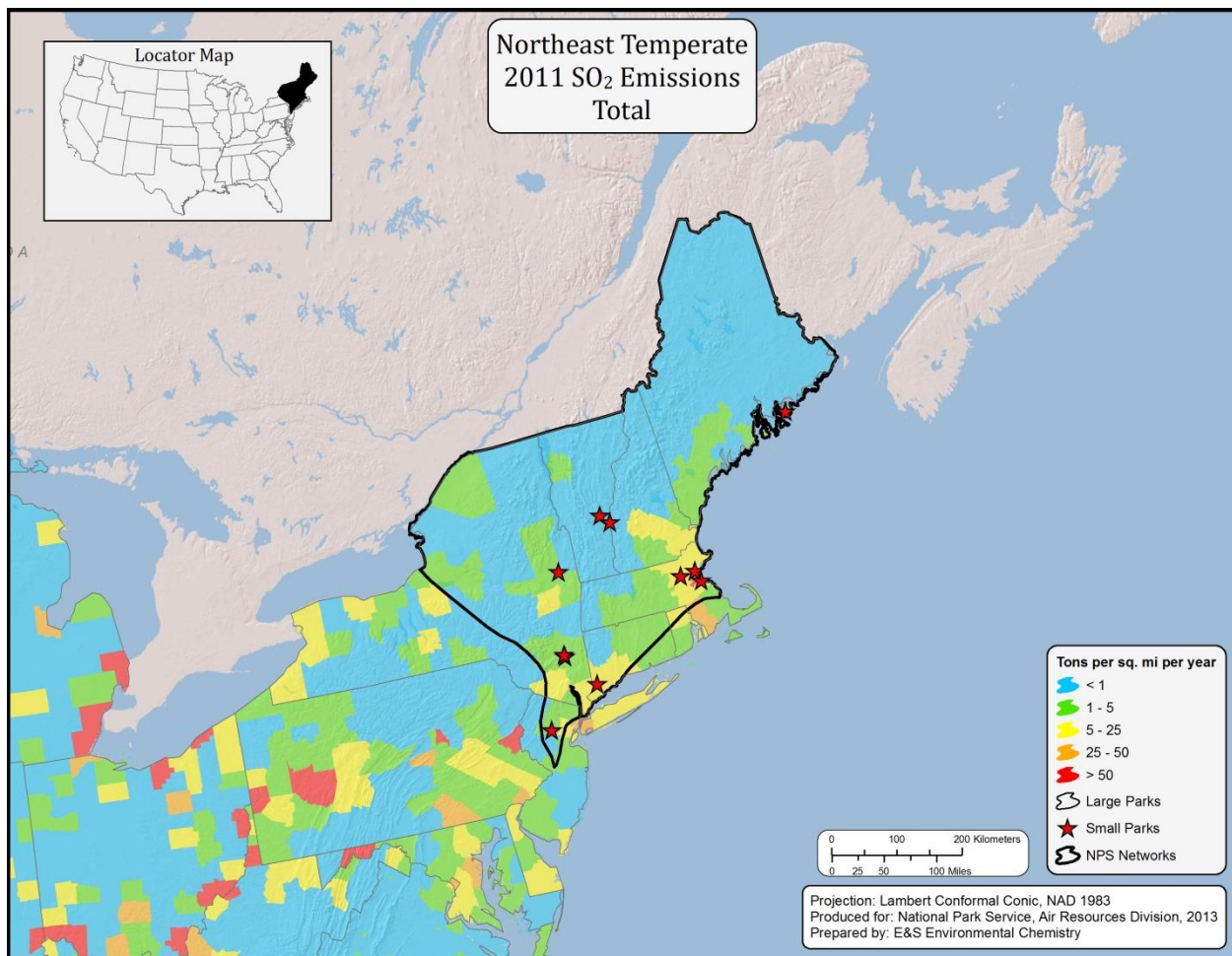
County-level emissions near NETN, based on data from the EPA's National Emissions Inventory (NEI) during a recent time period (2011), are depicted in Maps 2 through 4 for SO₂, oxidized N (NO_x), and reduced N (NH₃), respectively. Many counties to the southwest of NETN parks had relatively high SO₂ emissions (> 50 tons/mi²/yr; Map 2). Spatial patterns in NO_x emissions were generally similar, with highest values generally to the south and southwest of NETN parks (Map 3). Emissions of NH₃ near NETN parks were somewhat lower, with most counties showing emissions levels below 8 tons/mi²/yr (Map 4).

Some states have adopted emissions controls for toxic substances that go well beyond federal requirements (Smith and Trip 2005). In 1998, the New England Governors and Eastern Canadian Premiers (NEG-ECP) Mercury Action Plan (MAP) was adopted to regulate mercury (Hg) emissions and to control the use of Hg-containing products in the region. Aggressive emissions reduction goals were adopted by the New England states, Atlantic provinces, and Quebec. New York and New Jersey have also participated. This effort resulted in a reduction of Hg emissions by 55% in 2003 as compared with mid-1990 levels, and 2010 Hg emissions levels likely reflected reductions of over 80% (U.S. EPA 2010). These values are in line with the 1998 goals of the NEG-ECP MAP of at least 50% and 75% reductions by 2003 and 2010, respectively (Smith and Trip 2005).

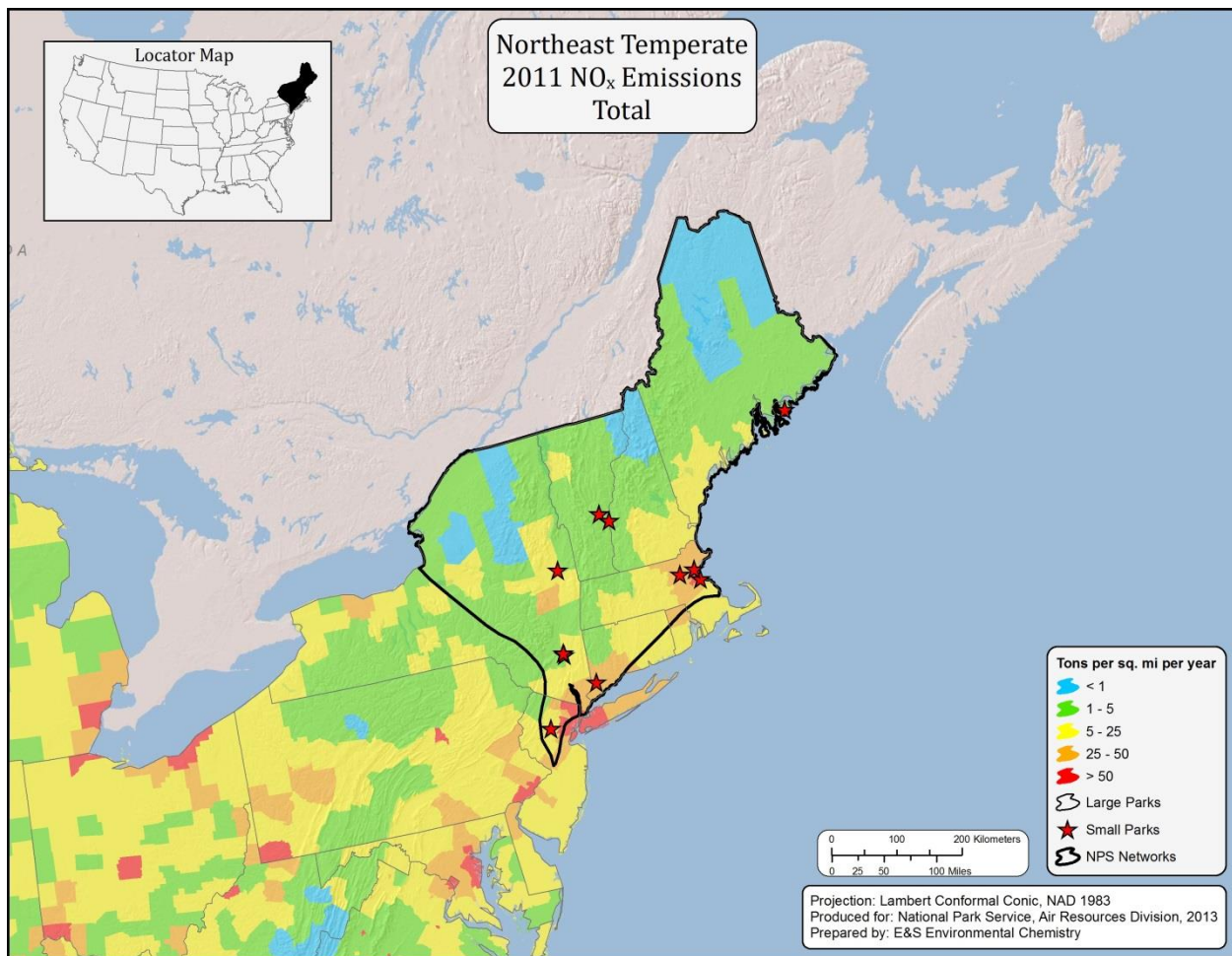
Total S and total N deposition within the NETN region in 2002 were both highly variable from north to south, ranging from between 2 and 5 kg S or N/ha/yr in northern Maine to between 10 and 15 kg S or N/ha/yr in much of the southern portion of the network region (Sullivan et al. 2011b). There were areas in the southern part of the network, especially around New York City, where estimates of both S and N deposition reached much higher levels.

Patterns of deposition have been extensively studied at ACAD. This park typically receives 20% to 40% more wet deposition of S than inland sites in Maine. During winter, air masses that reach ACAD tend to track over the ocean before reaching the park. Partly as a consequence of this pattern, mercury (Hg) and S wet deposition at ACAD are highest in summer and lowest in winter. Wet chloride (Cl⁻) deposition shows the opposite pattern (Kahl et al. 2007b).

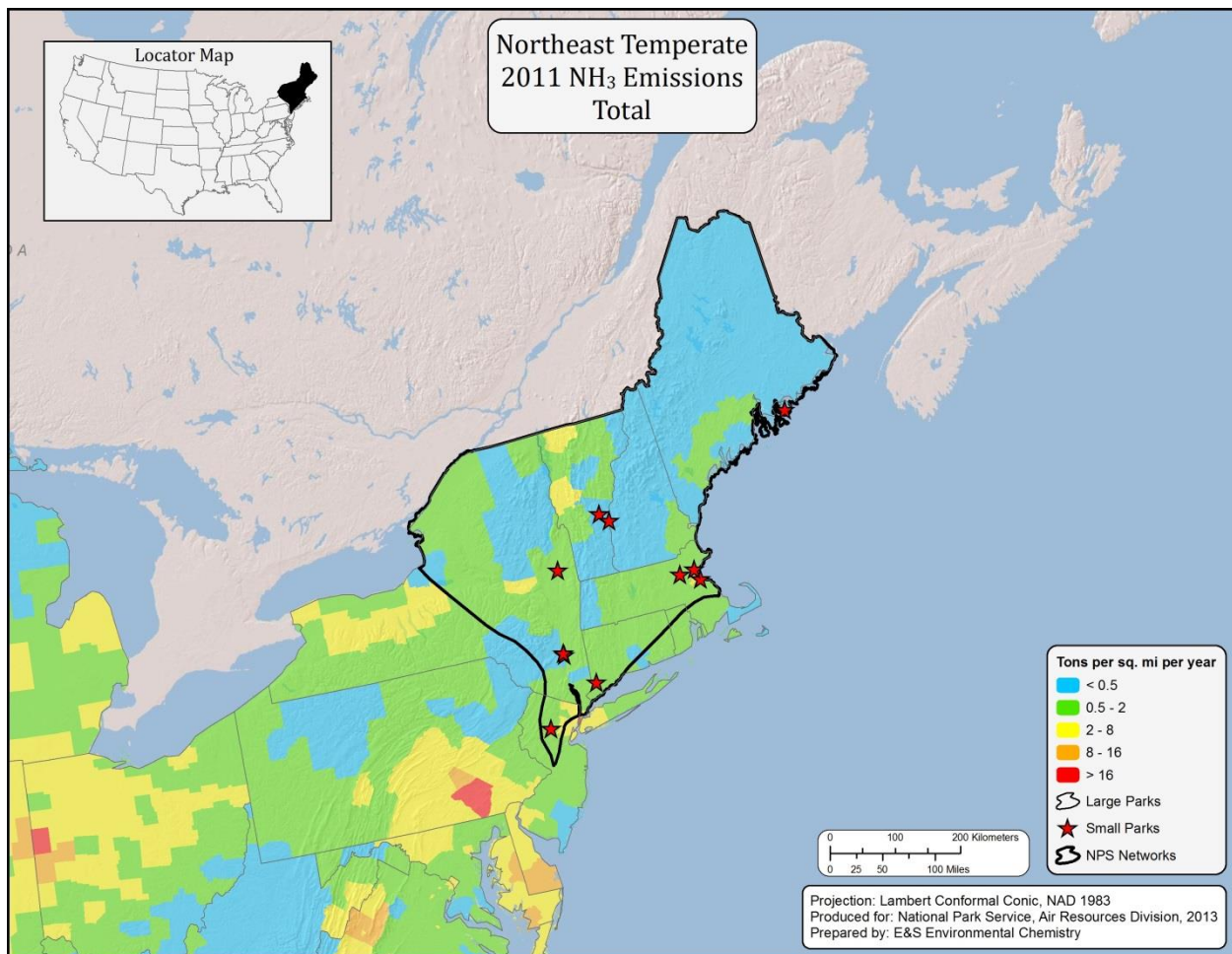
Dry deposition at ACAD varies with elevation and vegetation. The undisturbed coniferous forested landscape at ACAD contributes to high dry deposition of S, N, and Hg (Weathers et al. 2000, Weathers et al. 2006). Highest total S deposition occurs at high-elevation sites having coniferous forest vegetation. Weathers et al. (2006) simulated total S deposition hotspots in ACAD up to 25 kg S/ha/yr, and Kahl et al. (2007b) estimated maxima near 13 kg S/ha/yr. Results of both studies indicate that localized S deposition in parts of ACAD can be much higher than park or regional averages. Occult (fog and cloud) deposition can also be an important vehicle for depositing acidifying substances to ACAD and other coastal and high-elevation locations within the NETN. Fog



Map 2. Total SO₂ emissions, by county, near NETN for the year 2011. Data from EPA's National Emissions Inventory.



Map 3. Total NO_x emissions, by county, near NETN for the year 2011. Data from EPA's National Emissions Inventory.



Map 4. Total NH₃ emissions, by county, near NETN for the year 2011. Data from EPA's National Emissions Inventory.

pH below 3.5 has been documented in this park (Weathers et al. 1988a, Weathers et al. 1988b, Jagels et al. 1989).

Dry deposition of Hg at ACAD equals or exceeds wet deposition of Hg during both the growing season and the winter (Miller et al. 2005, Vaux et al. 2008). Snow Hg deposition is higher than previously thought (Nelson et al. 2007), but much of the Hg deposited in snow is subsequently volatilized and re-emitted back into the atmosphere (Vaux et al. 2008), limiting in-park environmental impacts.

Weathers et al. (2006) developed an empirical modeling approach, based on 300-400 throughfall measurements, to estimate total (wet, dry, plus cloud) deposition of S and N to the complex terrain of ACAD. Throughfall deposition measurements taken during summer, combined with landscape variables such as elevation, forest type, and slope, explained about 40% of the variation in total deposition estimates. Model estimates were scaled to wet and dry deposition measurements and estimated values from National Atmospheric Deposition Program (NADP) and Clean Air Status and Trends Network (CASTNET) monitoring sites. Resulting maps showed substantial spatial variability

in S and N deposition across the landscape of this park. Results of throughfall monitoring at 21 locations within ACAD indicated that canopy openness (which could also be represented as canopy height or vegetation type) and elevation were the most important landscape variables influencing spatial variability in total atmospheric deposition of S and Hg. The litterfall Hg flux was just as important as precipitation to the total input of Hg to the monitored sites. Elevated toxin deposition in ACAD has also been inferred from sediment accumulation rates of Hg and other contaminants (Kahl et al. 1985, Norton et al. 1997).

Model estimates of N loading to east coast, United States estuaries, including Casco Bay in southern Maine, have suggested that 15% to 42% of the total N loading was derived from atmospheric inputs (Castro and Driscoll 2002). These atmospheric inputs combine with substantial point and nonpoint non-atmospheric sources that also contribute N to estuaries in the NETN.

During the period 1985 to 2002, wet deposition of S decreased substantially at ACAD (Kahl et al. 2004), but there was not a significant change in NO_x or NH_x wet deposition (Lehmann et al. 2005). Because of the lack of improvement (decrease) in the levels of wet N deposition, Tonnessen and Manski (2007) recommend continued monitoring of ecosystems in ACAD to assess potential changes caused by fertilization and eutrophication.

Recently, Schwede and Lear (2014) documented a hybrid approach developed by the NADP Total Deposition (TDEP) Science Committee for estimating total N and S deposition. This approach combined monitoring and modeling data. Modeling was accomplished using the Community Multiscale Air Quality (CMAQ) model (Byun and Schere 2006). Priority was given to measured data near the locations of the monitors and to modeled data where monitoring data were not available. In addition, CMAQ data were used for N species that are not routinely measured in the monitoring programs: peroxyacetyl nitrate (PAN), N_2O_5 , NO, NO_2 , HONO, and organic NO_3 . The total deposition estimates are considered to be dynamic, with updates planned as new information becomes available. The TDEP data reported here were developed in late 2013 and are designated version 2013.02.

Atmospheric S deposition levels have decreased at all NETN parks since 2001, based on TDEP estimates (Table 1). Decreases in total S deposition over the previous decade for many parks in this network exceeded 50%. Estimated total N and oxidized N deposition over that same time period also decreased in all parks. Reduced N deposition was variable, decreasing in some parks and increasing in others.

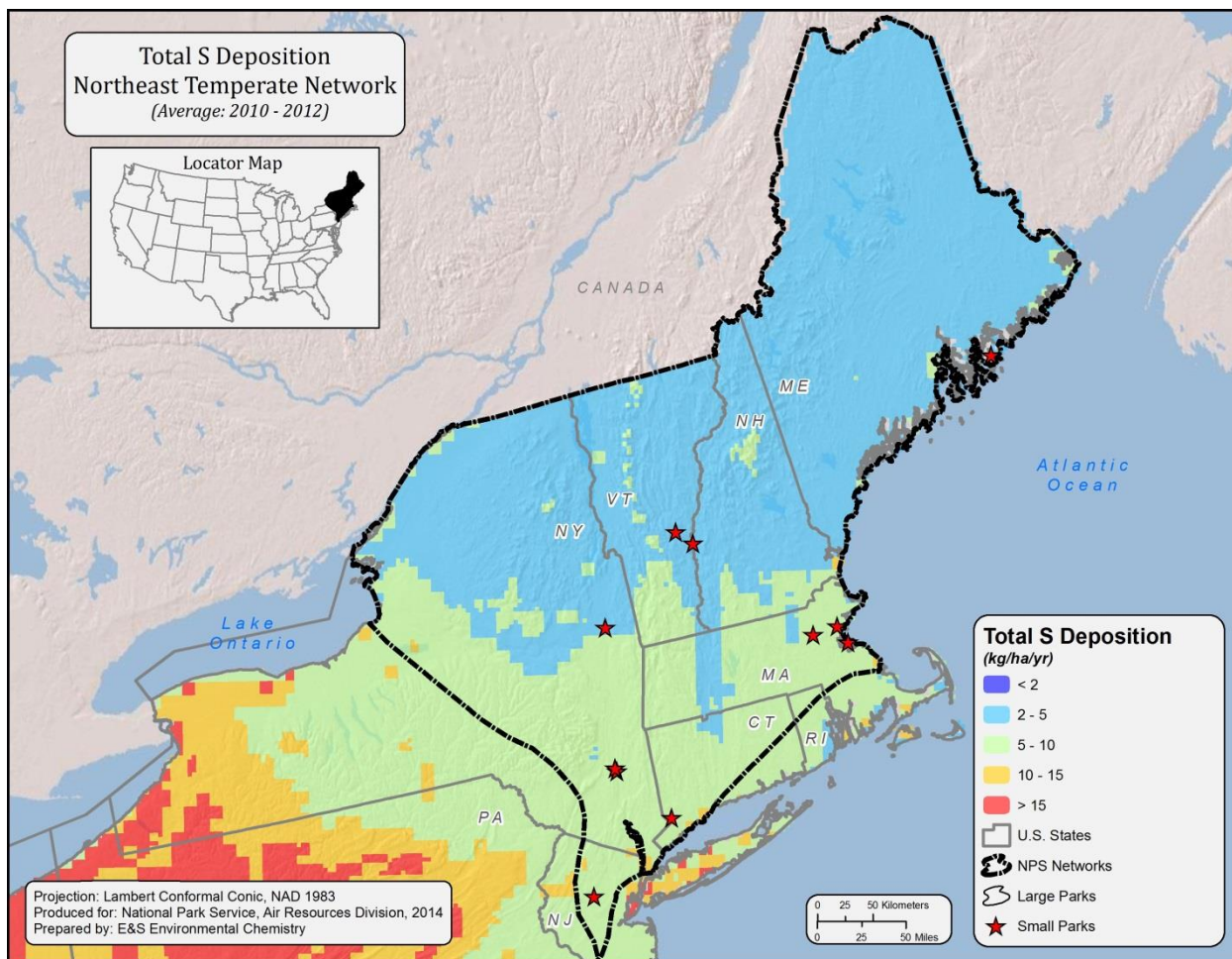
Total S deposition in and around NETN for the period 2010-2012 was generally highest ($> 5\text{-}10\text{ kg S/ha/yr}$) to the south and lowest ($< 5\text{ kg S/ha/yr}$) to the north of the network area (Map 5). Oxidized inorganic N deposition for the period 2010-2012 was in the range of $2\text{-}5\text{ kg N/ha/yr}$ throughout most of the park lands within NETN (Map 6). Estimates of NO_x deposition were higher to the south. Most areas received less than 5 kg N/ha/yr of reduced inorganic N from atmospheric deposition during this same period (Map 7). Total N deposition was higher than 10 kg N/ha/yr at some park locations, with highest values to the south (Map 8).

Table 1. Average changes in S and N deposition between 2001 and 2011 across park grid cells at NETN parks. Deposition estimates were determined by the Total Deposition Project, TDEP, based on three-year averages centered on 2001 and 2011 for all ~4 km grid cells in each park. The minimum, maximum, and range of 2011 S and N deposition within each park are also shown.

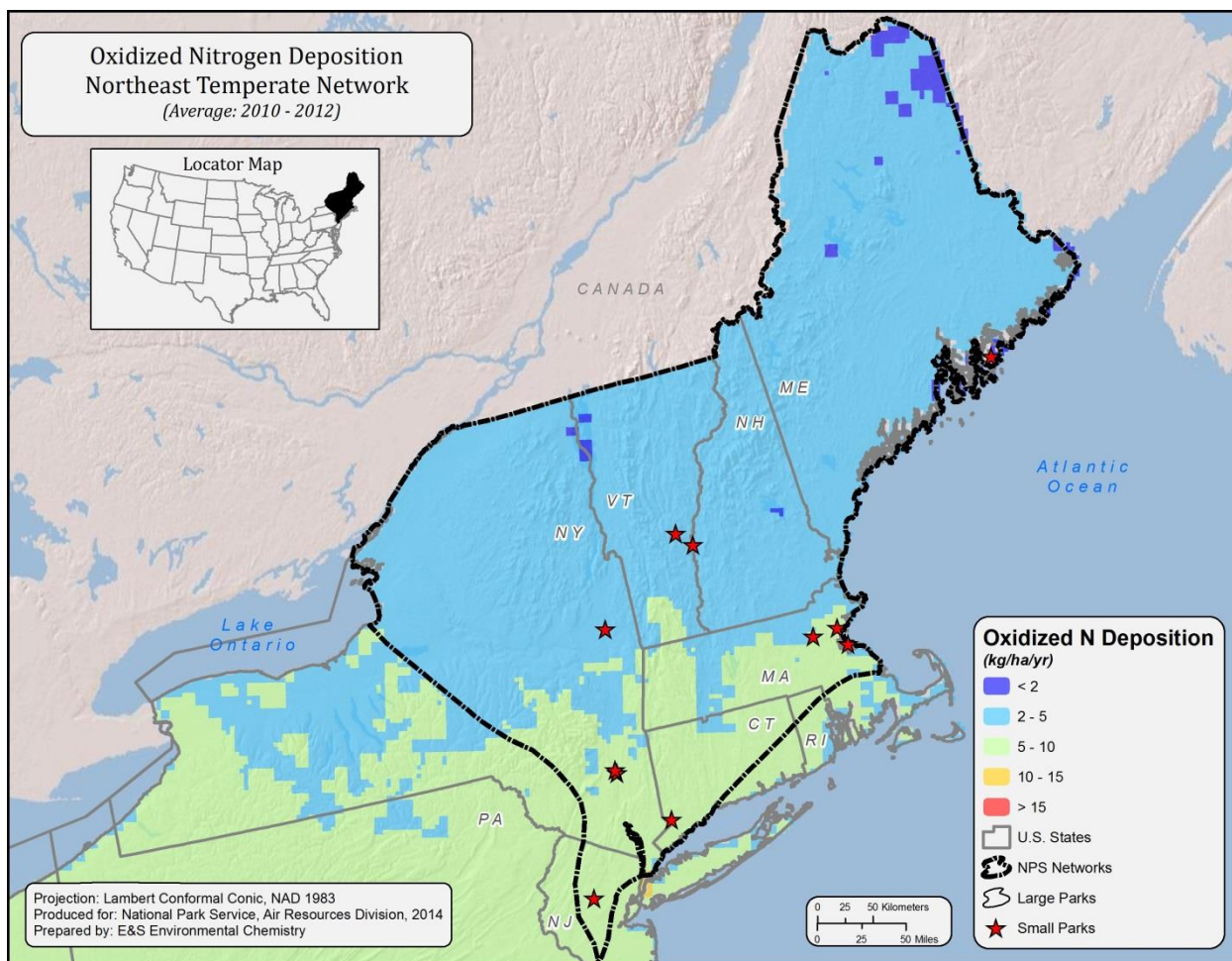
Park Code	Park Name	Parameter	2001 Average (kg/ha/yr)	2011 Average (kg/ha/yr)	Absolute Change (kg/ha/yr)	Percent Change	2011 Minimum (kg/ha/yr)	2011 Maximum (kg/ha/yr)	2011 Range (kg/ha/yr)
ACAD	Acadia	Total S	6.55	3.28	-3.28	-50.0%	3.14	5.66	2.52
		Total N	4.39	3.08	-1.31	-29.8%	2.97	5.17	2.20
		Oxidized N	3.06	1.82	-1.24	-40.4%	1.76	3.38	1.62
		Reduced N	1.33	1.26	-0.07	-5.5%	1.20	1.79	0.58
BOHA	Boston Harbor Islands	Total S	12.25	8.69	-3.56	-29.4%	6.06	9.86	3.81
		Total N	12.21	9.69	-2.52	-22.8%	4.99	11.77	6.78
		Oxidized N	8.76	5.58	-3.19	-37.3%	2.82	6.80	3.98
		Reduced N	3.44	4.11	0.66	16.5%	2.17	4.97	2.79
HOFR	Home of Franklin D. Roosevelt	Total S	12.75	6.11	-6.64	-52.0%	6.06	6.19	0.13
		Total N	11.58	8.97	-2.62	-22.6%	8.48	9.03	0.55
		Oxidized N	8.82	5.31	-3.51	-39.8%	5.16	5.34	0.18
		Reduced N	2.77	3.66	0.89	32.2%	3.33	3.69	0.37
MABI	Marsh-Billings-Rockefeller	Total S	7.97	3.77	-4.19	-52.6%	3.70	3.83	0.13
		Total N	8.78	5.77	-3.01	-34.3%	5.65	5.86	0.20
		Oxidized N	6.24	3.38	-2.85	-45.8%	3.33	3.42	0.09
		Reduced N	2.54	2.38	-0.16	-6.1%	2.32	2.43	0.11
MIMA	Minute Man	Total S	12.35	5.53	-6.82	-55.2%	5.50	5.57	0.08
		Total N	14.07	7.78	-6.30	-44.8%	7.61	7.85	0.25
		Oxidized N	10.42	5.16	-5.26	-50.5%	5.00	5.21	0.22
		Reduced N	3.65	2.62	-1.04	-28.4%	2.60	2.65	0.05
MORR	Morristown	Total S	16.39	9.09	-7.30	-44.5%	8.89	9.22	0.32
		Total N	19.00	12.67	-6.34	-33.4%	12.41	12.69	0.27
		Oxidized N	14.23	8.35	-5.88	-41.3%	8.27	8.37	0.11
		Reduced N	4.77	4.31	-0.46	-9.6%	4.15	4.35	0.21
SAGA	Saint-Gaudens	Total S	7.52	3.39	-4.13	-55.0%	3.26	3.58	0.32
		Total N	8.41	5.21	-3.20	-38.0%	5.10	5.36	0.25
		Oxidized N	6.08	3.04	-3.04	-50.0%	2.98	3.13	0.15
		Reduced N	2.33	2.17	-0.16	-6.8%	2.13	2.23	0.10

Table 1 (continued). Average changes in S and N deposition between 2001 and 2011 across park grid cells at NETN parks. Deposition estimates were determined by the Total Deposition Project, TDEP, based on three-year averages centered on 2001 and 2011 for all ~4 km grid cells in each park. The minimum, maximum, and range of 2011 S and N deposition within each park are also shown.

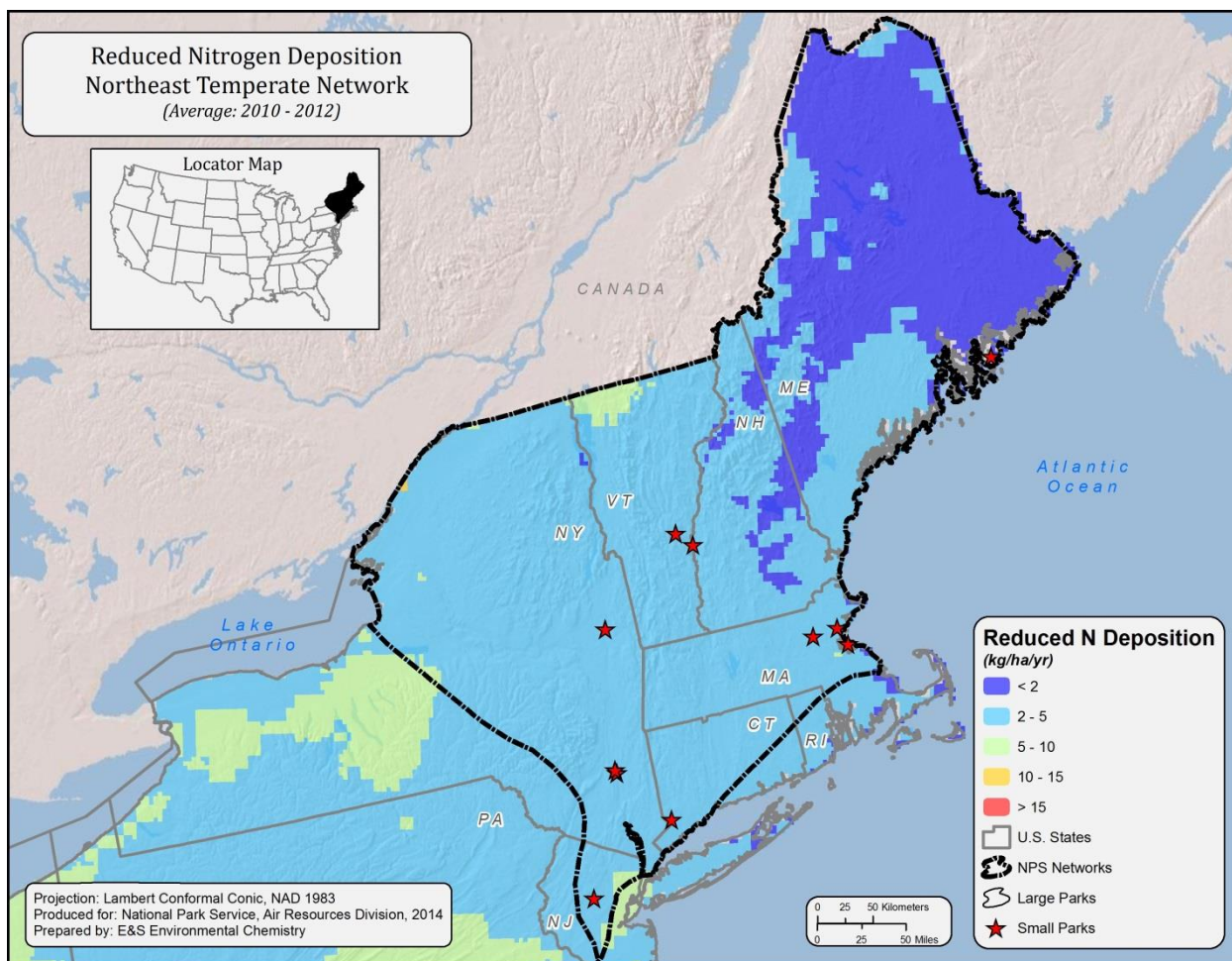
Park Code	Park Name	Parameter	2001 Average (kg/ha/yr)	2011 Average (kg/ha/yr)	Absolute Change (kg/ha/yr)	Percent Change	2011 Minimum (kg/ha/yr)	2011 Maximum (kg/ha/yr)	2011 Range (kg/ha/yr)
SAIR	Saugus Iron Works	Total S	11.87	7.21	-4.66	-39.3%	7.21	7.21	0.00
		Total N	13.76	8.16	-5.60	-40.7%	8.16	8.16	0.00
		Oxidized N	10.15	5.33	-4.82	-47.5%	5.33	5.33	0.00
		Reduced N	3.61	2.83	-0.78	-21.6%	2.83	2.83	0.00
SARA	Saratoga	Total S	11.06	4.78	-6.28	-56.8%	4.77	4.85	0.09
		Total N	12.04	6.84	-5.20	-43.2%	6.82	6.95	0.13
		Oxidized N	8.43	3.78	-4.65	-55.1%	3.77	3.84	0.06
		Reduced N	3.61	3.06	-0.56	-15.4%	3.05	3.12	0.07
VAMA	Vanderbilt Mansion	Total S	12.76	6.14	-6.62	-51.9%	6.12	6.15	0.03
		Total N	11.54	9.02	-2.51	-21.8%	9.00	9.03	0.04
		Oxidized N	8.77	5.33	-3.43	-39.2%	5.32	5.34	0.02
		Reduced N	2.77	3.69	0.92	33.2%	3.68	3.69	0.01
WEFA	Weir Farm	Total S	13.66	8.02	-5.64	-41.3%	8.02	8.03	0.01
		Total N	15.27	10.54	-4.72	-30.9%	10.52	10.64	0.12
		Oxidized N	12.02	7.01	-5.01	-41.7%	6.99	7.09	0.10
		Reduced N	3.25	3.53	0.28	8.7%	3.53	3.55	0.02



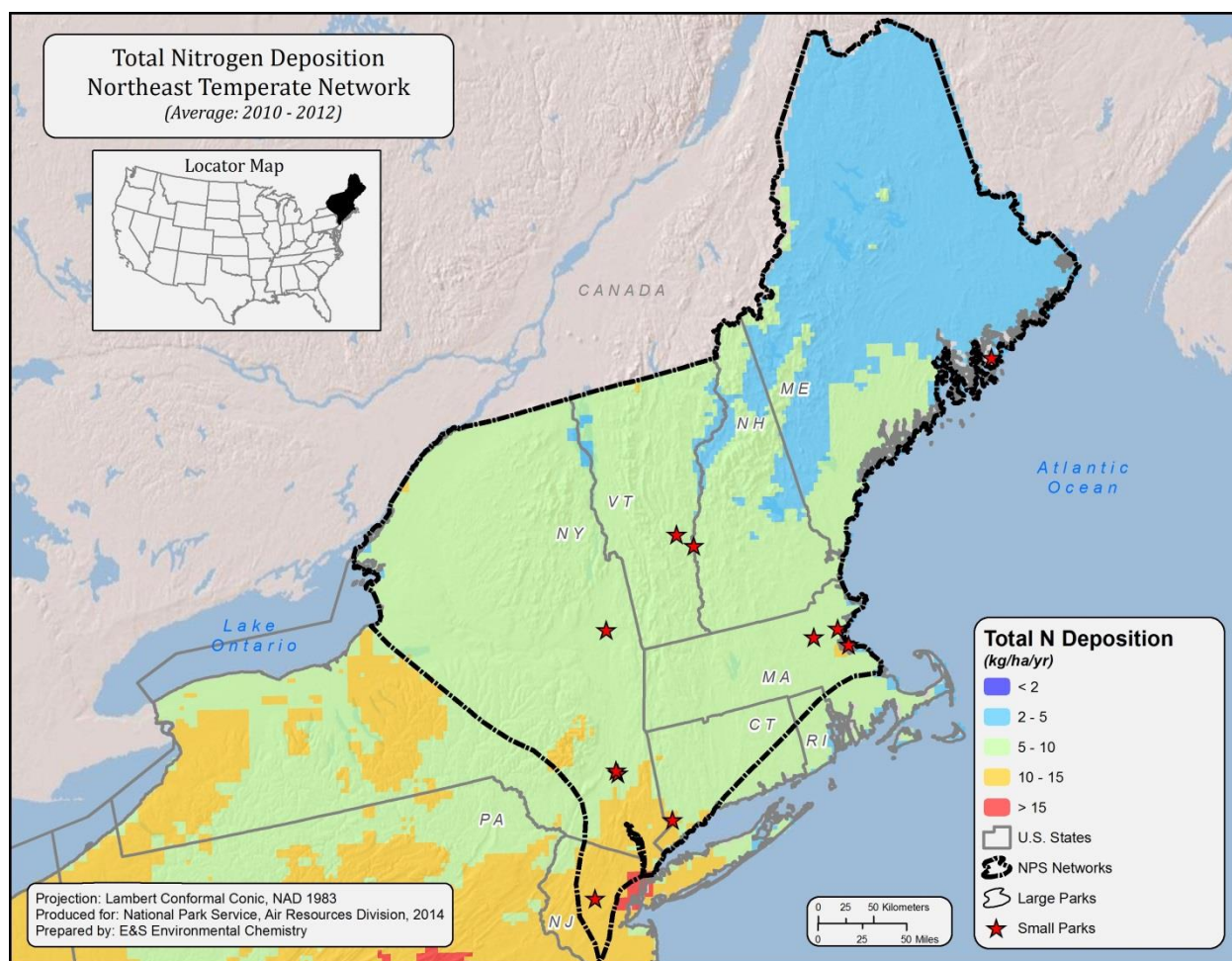
Map 5. Total S deposition for the three-year period centered on 2011 in and around NETN. (Source: Schwede and Lear 2014)



Map 6. Total oxidized inorganic N deposition for the three-year period centered on 2011 in and around NETN. (Source: Schwede and Lear 2014)



Map 7. Reduced inorganic N deposition for the three-year period centered on 2011 in and around NETN. (Source: Schwede and Lear 2014)



Map 8. Total N deposition for the three-year period centered on 2011 in and around NETN. (Source: Schwede and Lear 2014)

A study of two forested watersheds in ACAD by Johnson et al. (2007) investigated the effects of landscape factors, such as watershed aspect and forest type, on atmospheric Hg deposition. The coniferous forest study site received higher levels of total Hg (but not methylmercury [MeHg]) deposition compared with the site covered with hardwood vegetation (Johnson et al. 2007). The researchers interpreted this difference as likely due to the more efficient dry deposition scavenging of the conifer canopy which has waxy cuticle, greater surface roughness, higher foliar surface area, and denser canopy than a deciduous forest canopy (Johnson et al. 2007).

Litterfall can constitute an important depositional source of Hg to soils in the NETN. Sheehan et al. (2006) investigated the importance of Hg inputs to soil from litterfall in vegetated landscapes in ACAD. Results showed that Hg concentrations were significantly different among vegetation classes sampled. Highest concentrations were found in litter from softwoods (58.8 ± 3.3 ng Hg/g), followed by mixed (41.7 ± 2.8 ng Hg/g) and scrub (40.6 ± 2.7 ng Hg/g) vegetation types. Lowest Hg concentrations were found in litter from hardwoods (31.6 ± 2.6 ng Hg/g; Sheehan et al. 2006). There were no significant differences, however, among vegetation classes in litter Hg flux. This may have

been due to the high variability in the data and differences in litterfall mass between hardwoods and softwoods. Hardwood litter had higher autumnal Hg flux whereas softwood litter had higher Hg concentrations and higher litterfall flux throughout the winter and spring (Sheehan et al. 2006). Results suggested that the litterfall Hg flux can be just as large as the wet deposition flux.

Snow is a factor to consider when determining atmospheric Hg flux to soil, but has not been well-studied in the NETN. To address this gap in understanding, a study by Nelson et al. (2008) in ACAD examined Hg concentrations in snow and compared concentrations at sites with and without substantial conifer forest canopies, using differing collection methods. Results showed that Hg deposition at sites with no canopy, including the Mercury Deposition Network (MDN) site, was approximately 3.4 times lower than deposition at forested sites, as measured after snowfall “events” (snowfall of >8 cm). Mercury concentrations were lowest when measured as season-long throughfall Hg flux ($1.8 \mu\text{g}/\text{m}^2$), slightly higher in the bulk snowpack ($2.38 \pm 0.68 \mu\text{g}/\text{m}^2$), and highest in measurements made after snowfall events ($5.63 \pm 0.38 \mu\text{g}/\text{m}^2$; Nelson et al. 2008). These results illustrate the importance of dry deposition during the cold season, and subsequent re-emission of Hg from the snowpack back into the atmosphere, to the annual Hg flux from the atmosphere to the soil. Additionally, the researchers conducted an Hg tracer study and found evidence for movement of Hg from the soil into the snowpack (Nelson et al. 2008).

Acidification

The network rankings constructed by Sullivan et al. (2011b) in a coarse screening study for acid Pollutant Exposure, Ecosystem Sensitivity to acidification, and Park Protection yielded an overall network acidification Summary Risk ranking for the NETN that was near the middle of the distribution among networks. While rankings are an indication of risk, park-specific data, particularly data on ecosystem sensitivity, are needed to fully evaluate risk from acidification.

Acid Pollutant Exposure rankings for the individual parks in the NETN (Table 2) were variable, from Moderate (middle quintile) for four of the parks (including ACAD) to Very High (highest quintile) for six parks. Acid sensitivity varied also, from Low to Very High, depending on landscape characterization. ACAD's steep slopes, thin soils, high mountains, and exposure to coastal fog increase the park's susceptibility to deposition of long-range atmospheric pollutants, including S, N, mercury (Hg), and other contaminants (Kahl et al. 2000). The three NETN parks located furthest north (ACAD, MABI, SAGA) were all ranked Very High for ecosystem sensitivity to acidification. Most of the other parks in the network were ranked High or Moderate in acid sensitivity.

Previous research on acidification in northeastern national parks has focused on ACAD. A pair of gauged watershed research sites was established at ACAD in 1990, as part of the Park Research and Intensive Monitoring of Ecosystems Network (PRIMENet), a joint program of EPA and NPS. Research has focused on quantifying the influence of landscape on atmospheric S, N, and Hg deposition; quantifying throughfall fluxes of Hg; investigating seasonal patterns of deposition; and documenting relationships between atmospheric deposition and stream chemistry (Kahl et al. 2007b).

The Ecosystem Sensitivity to acidification rankings reported by Sullivan et al. (2011b) were variable among parks in the NETN, from Low for one of the parks (BOHA) to Very High for four of the parks (including ACAD; Table 2). All of the streams in ACAD are first through third order. Such streams tend to be more likely to be sensitive to acidification than larger, higher-order streams.

Table 2. Estimated I&M park rankings¹ according to risk of acidification impacts on sensitive receptors. (Source: Sullivan et al. 2011b)

Park Name	Park Code	Estimated Acid Pollutant Exposure	Estimated Ecosystem Sensitivity to Acidification
Acadia	ACAD	Moderate	Very High
Boston Harbor Islands	BOHA	Very High	Low
Home of Franklin D. Roosevelt	HOFR	Very High	Moderate
Marsh-Billings-Rockefeller	MABI	Moderate	Very High
Minute Man	MIMA	High	High
Morristown	MORR	Very High	Very High
Saint-Gaudens	SAGA	Moderate	Very High
Saratoga	SARA	Moderate	Moderate
Saugus Iron Works	SAIR	Very High	High
Vanderbilt Mansion	VAMA	Very High	Moderate
Weir Farm	WEFA	Very High	High

¹ Relative park rankings are designated according to quintile ranking, among all I&M Parks, from the lowest quintile (Very Low risk) to the highest quintile (Very High risk).

Ecosystem sensitivity to acidification rankings also take into account land slope, which often influences the degree of acid neutralization provided by soils and bedrock within the watershed. Six of the parks in the NETN have low relief, with less than 10° average slope. The remaining five parks have average slope between 10° and 20°. Topography in ACAD ranges from rolling hills to mountains.

Most of ACAD is underlain by granite. The low buffering capacity of the overlying soils, together with areas having steep slopes, and in some areas also relatively high elevation, influence susceptibility of soil and drainage water to acidification (Kahl et al. 2000). Rainfall in ACAD is about 140 cm/yr, which is about 20 cm/yr higher than the rest of Maine (Nielsen and Kahl 2007). These features enhance the sensitivity of streams and soils in the park to acidification. The predominant sensitive vegetation type in ACAD is red spruce. There is also some sugar maple present. These species are well known to be acid-sensitive.

The NETN has conducted forest health monitoring in ACAD since 2006. Nearly all of the 176 forest health monitoring plots have been sampled twice through 2013 (Miller et al. 2014). Overall forest health in ACAD is good, although the forests are subjected to multiple stressors, including soil acidification. Some forest soils in ACAD have high organic content, low pH, and low base saturation (Miller et al. 2014).

Acidification of Terrestrial Ecosystems

Acid-sensitive forest soils have been acidified by sulfur oxides (SO_x) and NO_x deposition throughout the NETN region. However, much of the available information for the NETN region has been collected in ACAD, in the Adirondack and Catskill mountains to the northwest of the NETN parks, or in the national forests of New Hampshire and Vermont. Acidic deposition has been shown to be an important factor causing decreases in concentrations of exchangeable base cations in soils which

were naturally low historically throughout much of the NETN region. Base saturation values less than 10% predominate in the B horizon in areas where soil and surface water acidification from acidic deposition have been most pronounced (David and Lawrence 1996, Bailey et al. 2004, Sullivan et al. 2006a). At Hubbard Brook Experimental Forest (HBEF) in New Hampshire, model hindcast simulations suggest that soil percent base saturation has decreased due to acidic deposition and accumulation of nutrient cations by forest vegetation. Sulfur and N emissions controls put into place in the United States (and neighboring Canada) since passage of the CAA and its Amendments (CAAA) have slowed the pace of damage to sensitive ecosystems in the NETN region.

Forest soils in ACAD, which have high organic content, low pH, and low base saturation, are most vulnerable to acidification among the parks in NETN, despite receiving the lowest annual acid deposition rates in the network (Miller et al. 2012). Acidification risk is low in the well-buffered soils in MABI, MORR and SARA and varies in the remaining parks. Some monitoring plots in SAGA, MIMA and ROVA are comprised of acidic, poorly-buffered soils, and other plots appear to be well-buffered (Miller et al. 2012).

To some degree, soils can recover their base cation reserves over time in response to reduced levels of acidic deposition. Some degree of soil and/or vegetation recovery may be beginning to occur in some places in the NETN as air pollution levels decline, but scientists have not yet been able to confirm that response. It is unlikely that recovery from soil damage has occurred to any significant extent in the Northeast in response to decreased levels of air pollution. More likely, soil conditions and plant health are continuing to decline, albeit more slowly, at most acid-sensitive locations. The recovery potential of soil exchangeable base cation concentrations is dependent on weathering rates, which can be very slow. Additionally, there is evidence of a delayed watershed acidification recovery response. In the past, most of the S deposited in watersheds in the NETN region moved more or less directly through soils and into water as SO_4^{2-} , with the capacity to acidify soil and water along the way. Nevertheless, a fraction of the S deposited each year was stored in the soil. Now that S deposition inputs to the watersheds have decreased due to air pollution regulations, some of that stored S is being released to drainage water, making recovery slower than it would be otherwise as a consequence of the cumulative damage to the soil from past air pollution. Current scientific understanding suggests that additional cuts in emissions, beyond those required as of about 2003, might enable the most sensitive ecosystems to continue to recover, and prevent renewed acidification in response to base cation loss from soils under continued (albeit lower) levels of atmospheric S and N deposition (Sullivan et al. 2006b).

Acidic deposition and soil acidification can have detrimental effects on multiple forest species. It is difficult to quantify these effects, in part because plants are simultaneously being affected by multiple stressors; besides air pollution, plants are particularly affected by changing climatic conditions, insect pests, and disease. Nevertheless, it is clear that plant species throughout portions of the NETN region have been damaged by air pollution. At sensitive locations, acidic deposition has resulted in base cation depletion and decreased Ca:Al ratio in soil solution. Aluminum (Al) is present naturally in soils, where it occurs mostly in solid form. However, under high levels of acidic deposition, at locations where the soil base cation supplies are limited, some soil Al becomes

dissolved in soil water. Dissolved Al is toxic to plant roots, fish, and other life forms. Additionally, Aber et al. (2003) concluded that N deposition is altering the N status of northeastern forests; the study reported a decrease in C:N ratio from about 35 to about 25 along an increasing atmospheric N deposition gradient of 3 to 12 kg N/ha/yr across the Northeast.

Soil acidification, Al toxicity, and exposure of foliage to acidic deposition have collectively contributed to decline in some tree species in portions of the northeastern United States that have experienced soil acidification as a consequence of SO_x and NO_x deposition. Effects have included reduced growth and increased stress to overstory trees, and likely changes in the species distributions of understory plants. In the northeastern United States, two species of coniferous tree (red spruce and red pine [*Pinus resinosa*]) have been shown to be especially sensitive to acidification. Both occur at many of the NETN parks. Acidic deposition has been implicated as a causal factor for the decline of red spruce at high elevation throughout the NETN region (DeHayes et al. 1999). Spruce dieback has been observed and has been most severe at high elevations in the Adirondack and Green Mountains, where more than 50% of the canopy trees died during the 1970s and 1980s. In the White Mountains, about 25% of the canopy spruce died during that same period (Craig and Friedland 1991). Dieback of red spruce trees was also observed in mixed hardwood-conifer stands at relatively low elevations in the western Adirondack Mountains (Shortle et al. 1997). This rapid dieback was linked with two aspects of air pollution: exposure of foliage to acidic cloud water and an increase in the amount of dissolved Al compared with dissolved Ca in soil water. The extent to which such effects have occurred in the national parks of the NETN is not known.

Some of the red spruce decline in the NETN region was documented at high-elevation sites which frequently experience cloud cover. Much of the total atmospheric N deposition load at such locations likely comes in the form of cloud deposition, which is often more acidic than acid rain. Results of controlled exposure studies showed that acidic mist or cloud water reduced the cold tolerance of current-year red spruce needles by 3 to 10 °C (DeHayes et al. 1999), and this response may be at least partially responsible for the observed dieback. The frequency of freezing injury of red spruce has increased over the past half century, a period that coincided with increased emissions of S and N oxides and increased acidic deposition (DeHayes et al. 1999). There is a significant positive association between cold tolerance and foliar Ca in trees that exhibit deficiency in foliar Ca. The membrane-associated pool of Ca²⁺, although a relatively small fraction of the total foliar Ca pool, strongly influences the response of cells to changing environmental conditions. The plant plasma membrane plays an important role in mediating cold acclimation and low-temperature injury (U.S. EPA 2004). The studies of DeHayes et al. (1999) suggested that the direct deposition of acidic compounds on red spruce needles preferentially removes membrane-associated Ca. More recently a strong link has also been established between availability of soil Ca²⁺ and winter injury (Hawley et al. 2006).

Experimental red spruce plots on Mount Ascutney, Vermont were fertilized with N in order to evaluate effects on foliar membrane Ca, winter cold tolerance, and freezing injury. Each of six plots in a red spruce stand received treatments of 0, 15.7, or 31.4 kg N/ha/yr for 12 years. Foliage from trees in plots receiving N additions showed lower cold tolerance, greater freezing injury, membrane-

associated Ca deficiencies, and higher electrolyte leakage relative to controls (McNulty and Aber 1993, McNulty et al. 1996, Schaberg et al. 2000, 2002).

Thus, the weight of evidence suggests the likelihood of adverse impacts of acidification from atmospheric S and N deposition on coniferous forests in the Northeast and, possibly, at NETN parks. Nevertheless, other than in ACAD, park-specific data are generally lacking in this network.

Acidification can also affect the base saturation, availability, and leaching loss of important base cation nutrients in hardwood forests. Deciduous forests show variable responses to acidification depending on the tree species present. The deciduous tree species most commonly associated with adverse acidification-related effects of S and N deposition is sugar maple; it occurs in the NETN parks. Sugar maple is a key species of the northern hardwood forest that predominates in many portions of the northeastern United States and central Appalachian Mountain region. Several studies, mainly in Pennsylvania (Bailey et al. 2004) and the Adirondack Mountains (Sullivan et al. 2013), have indicated that sugar maple decline is linked to the occurrence of relatively high levels of acidic deposition and base-poor soils.

Along an increasing N deposition gradient in the northeastern United States, from 4.2 to 11.1 kg N/ha/yr, Lovett and Rueth (1999) found a twofold increase in N mineralization in soils of sugar maple stands, but no significant relationship between increased deposition and mineralization in American beech (*Fagus grandifolia*) stands. This difference might be attributable to the lower litter quality in beech stands. Sugar maple appears to be more susceptible to effects of increasing deposition and concomitant soil acidification from either direct leaching of NO_3^- or enhanced nitrification. For northeastern hardwoods, Aber et al. (2003) found a decrease in soil C:N ratio from 24 to 17 over a deposition gradient of 3 to 12 kg N/ha/yr. This decrease was similar to, but less steep than, the decrease seen in soil under conifers.

The health of sugar maple trees is strongly influenced by the availability of Ca in soil. Calcium is depleted by acidic deposition. Trees that grow on soils having low base cation supply are stressed and consequently often become more susceptible to damage from defoliating insects, drought, and extreme weather. The overall response includes death of mature trees and poor regeneration of seedlings. In a study of sugar maple decline throughout the Northeast, Bailey et al. (2004) found threshold relationships between base cation availability in the upper B soil horizon and sugar maple mortality at Ca^{2+} saturation less than 2%, and magnesium (Mg^{2+}) saturation less than 0.5%.

Stand age and successional stage also can affect the susceptibility of hardwood forests to acidification effects. In northeastern hardwood forests, older stands exhibit greater potential for Ca^{2+} depletion in response to acidic deposition than younger stands. Thus, with the successional change from pin cherry (*Prunus pensylvanica*), striped maple (*Acer pensylvanicum*), white ash (*Fraxinus americana*), yellow birch and white birch (*Betula papyrifera*) in younger stands to American beech and red maple (*Acer rubrum*) in older stands, there is an increase in sensitivity to acidification (Hamburg et al. 2003).

Across an 800 km pollution gradient (3 to 11 kg SO₄-S/ha/yr; 2 to 4 kg NO₃-N/ha/yr) in northern hardwood forests, with maples dominant, Pregitzer and Burton (1992) found a 200 to 300 µg/g increase in foliar S, and litter fall S content ranged from 872 to 1356 µg/g. While foliar N did not change across the gradient, litterfall N was correlated with changing deposition. Pregitzer and Burton (1992) asserted that their data did not suggest a causal link between acidic deposition and forest decline. Decline would be impossible to document in the short five-year time frame of their study. They did, however, assert that their results supported the plausibility of altered tree nutrition across large geographic regions in NETN due to acidic deposition.

Acidification of Aquatic Ecosystems

Status

The high rates of atmospheric deposition of S and N on the watersheds of lakes and streams in the NETN region, combined with naturally low contributions from some rock types of base cations that serve to neutralize acidity, are among the most important causes of acidity in many lakes and streams within this region.

Sulfur deposition has contributed to chronic surface water acidification in the northeastern United States to a greater extent than has N deposition. Nitrate concentrations in acid-sensitive drainage waters are generally much lower than SO₄²⁻ concentrations. However, at the peak of snowmelt, the influence of N deposition becomes proportionately more important, and in some waters is just as important as S acidity. The seasonal shift in the relative importance of S- and N-caused acidity is related to the dynamics of plant and microbial growth cycles and snowpack accumulation and melting.

Most lakes in ACAD are circumneutral, with average pH between 6.5 and 7.5. Only two are known to have had pH < 5.0: Duck Pond is naturally acidic from organic acids; Sargent Mountain Pond has limited soil in its watershed and has likely been acidified by acidic deposition (Kahl et al. 2000, Vaux et al. 2008). Most surveyed streams in the park have acid neutralizing capacity (ANC) < 100 µeq/L, and seven surveyed streams were acidic at the time of sampling (ANC ≤ 0 µeq/L; Vaux et al. 2008). The relatively recent NETN monitoring has not revealed any acidic lakes in ACAD. Kahl et al. (2007b) surveyed 28 streams in ACAD. Streamwater SO₄²⁻ concentrations followed the observed spatial patterns in S deposition. Deposition and streamwater SO₄²⁻ concentration were both highest in the western and southern sections of the park. However, the relationship between Hg deposition and streamwater Hg concentration was more complex.

Data from over 12,500 streams and lakes were used by the Critical Loads of Atmospheric Deposition Science Committee (CLAD) of the NADP (<http://nadp.sws.uiuc.edu/committees/clad/>) to develop steady-state critical loads for acidity of surface waters based on multiple approaches for estimating base cation weathering; modified F-factor, regional regression model, and MAGIC model. Water quality data were obtained from a variety of sources including EPA Long Term Monitoring (LTM) sites, lake surveys, EMAP Assessments, and National Stream Surveys; USGS; NPS Vital Signs program; and USFS air program. The average water quality measurements from the most recent five years of data were used for sites with long-term water monitoring records. The CLAD database

included 2 sites in MABI, 3 sites in MORR, and 85 sites in ACAD. Streams in ACAD has modeled CL as low as 0 eq S/ha/yr, but all modeled values at the other two parks were high (> 3000 eq S/ha/yr).

Past Trends

Over the past 25 years, S deposition to acid-sensitive watersheds in the NETN has decreased by about half. The pH of rainfall has increased by about a fourth of a pH unit, and N deposition has decreased slightly.

EPA's Long Term Monitoring (LTM) Program has been collecting monitoring data since the early 1980s for many lakes and streams in acid-sensitive areas of the United States, including within the NETN region. These data allow evaluation of trends and variability in key components of lake and streamwater chemistry prior to, during, and subsequent to CAAA Title IV implementation. Throughout the NETN, the concentration of SO_4^{2-} in surface waters has decreased substantially, often by a third or more, subsequent to the CAAA. Monitoring data from the LTM and TIME projects indicated that most regions included in the monitoring efforts showed large declines in SO_4^{2-} concentrations in surface waters over the period of monitoring (1990s) analyzed by Stoddard et al. (2003), with rates of change for individual lakes ranging from about -1.5 to -3 $\mu\text{eq/L/yr}$. These declines in lake and stream SO_4^{2-} concentrations were considered consistent with observed declines in S wet deposition, and were corroborated by other studies that showed that SO_4^{2-} concentrations in Adirondack lakes have decreased steadily since at least 1978 (e.g., Driscoll et al. 1995, Stoddard et al. 2003).

Trend analysis results for the period 1982 to 1994 were reported by Stoddard et al. (1998) for 36 lakes in the northeastern United States having $\text{ANC} \leq 100 \mu\text{eq/L}$. Trend statistics among sites were combined through a meta-analytical technique to determine whether the combined results from multiple sites had more significance than the individual Seasonal Kendall Test statistics. All lakes showed significant declining trends in SO_4^{2-} concentration ($\Delta \text{SO}_4^{2-} = -1.7 \mu\text{eq/L/yr}$; $p \leq 0.001$). Lakewater ANC responses were regionally variable. Lakes in New England showed evidence of ANC recovery ($\Delta \text{ANC} = 0.8 \mu\text{eq/L/yr}$; $p \leq 0.001$), whereas Adirondack lakes exhibited either no trend or further acidification during that time period (through 1994), largely because of declines in base cation concentrations. Recovery of Adirondack lake ANC became evident in more recent times. Lake ANC increased in the Adirondacks during the decade of the 1990s at a rate of about +1 $\mu\text{eq/L/yr}$, despite a continued decline in surface water base cation concentrations; Adirondack lake Al_i concentrations declined slightly. Regional surface water ANC did not change significantly in New England during that time period (Stoddard et al. 2003).

There were no significant changes in the ANC or SO_4^{2-} concentration in streams in ACAD during the period 1982 to 1990 (Heath et al. 1993). During the period 1990 to 2000, however, the concentrations of SO_4^{2-} and base cations in lakes in ACAD decreased by significant amounts (10% decrease for SO_4^{2-}), but the pH did not increase (Kahl et al. 2004).

It was hoped that emissions reductions required by the CAAA would result in recovery of lake and stream chemistry, including increases in pH and ANC values and decreases in Al_i concentrations in

surface water. Such improvements in water chemistry would, in turn, benefit acid-sensitive fish, invertebrates, and other life forms. It was also hoped that damaged soils and plant species would show signs of recovery. Hopes for recovery have varied among scientists. The prevailing expectation has shifted from the rapid recovery generally expected during the 1980s to the prevailing current expectation of a long-term (decades or longer) gradual process of chemical recovery.

Insights can be gained from site-specific long-term studies in the NETN region. The HBEF has one of the longest continuous records of precipitation and streamwater chemistry in the United States. Long-term stream water data from the HBEF, which is located near MABI and SAGA, revealed a number of changes that are consistent with trends observed in lakes and streams across eastern North America (Stoddard et al. 1999, Evans and Monteith 2001, Stoddard et al. 2003). Stream water draining the HBEF reference watershed (Watershed 6) had a 32% decline in the annual volume-weighted concentration of SO_4^{2-} ($-1.1 \mu\text{eq/L/yr}$) between 1963 and 2000 (Driscoll et al. 2007a). This decrease in stream SO_4^{2-} concentration was related to decreases in atmospheric emissions of SO_2 and to bulk precipitation concentrations of SO_4^{2-} (Likens et al. 2001). In addition, there was a long-term decrease in stream concentrations of NO_3^- that was not correlated with a commensurate change in emissions of NO_x or in bulk deposition of NO_3^- . The long-term declines in stream concentrations of strong acids ($\text{SO}_4^{2-} + \text{NO}_3^-$; $-1.9 \mu\text{eq/L/yr}$) resulted in small but significant increases in pH, from 4.8 to 5.0 (Driscoll et al. 2007a). The pH of streams at HBEF remain low compared to background conditions, estimated to be approximately pH = 6.0 (Driscoll et al. 2007a). The recent increase in stream pH was limited because of concurrent decreases in the sum of base cations ($-1.6 \mu\text{eq/L/yr}$; Driscoll et al. 2001). Driscoll et al. (2001) estimated that roughly 6% of lakes and streams in the Northeast are considered more sensitive to acidic deposition than the reference stream monitored at HBEF (Driscoll et al. 2001).

Mitchell and Likens (2011) analyzed long-term (1965-2008) S budget data for several watersheds in HBEF. They documented increasing importance of S release from internal storage pools over time. Watershed wetness, as a function of $\log_{10}$ annual water flux, explained 57% of the annual variation in SO_4^{2-} export. Mitchell and Likens (2011) argued that biogeochemical control of SO_4^{2-} export in stream water of forested watersheds in HBEF has shifted from atmospheric S deposition to climatic factors that affect soil moisture.

The observed changes in the concentration of NO_3^- in some surface waters in the NETN region have likely been due to a variety of factors, including climate. During the 1980s, NO_3^- concentration increased in many surface waters (Driscoll and Van Dreaseon 1993, Murdoch and Stoddard 1993). There was concern that some forests were becoming N-saturated, leading to increased NO_3^- leaching from forest soils throughout the region. Such a response could partially negate the benefits of decreased SO_4^{2-} concentrations in lake and stream waters. However, this trend was reversed in about 1990, and the reversal could not be attributed to a change in N deposition. Nitrate leaching through soils to drainage waters is the result of a complex set of biological and hydrological processes. Key components include N uptake by plants and microbes, transformations between the various forms of inorganic and organic N, and local precipitation patterns. Most of the major processes are influenced by climatic factors, including temperature, moisture, and snowpack development. Therefore, NO_3^-

concentrations in surface waters respond to many factors in addition to N deposition and can be difficult to predict. It is likely that monitoring programs of several decades or longer will be needed to separate trends in NO_3^- leaching from climatic variability in forested watersheds in the NETN.

Lakewater SO_4^{2-} concentrations in the most acid-sensitive Maine lakes declined by about 12% to 22% during the period 1982 to 1998 (Kahl 1999). Only in the seepage lakes, however, was there evidence of a small decline in lakewater acidity during that period. The high-elevation lakes in Maine showed small declines in lakewater acidity during the 1980s, but that trend slowed or reversed in the 1990s (Kahl 1999). Whereas NO_3^- concentrations decreased during the 1990s in many lake chemistry datasets (cf., Stoddard et al. 2003), the high-elevation lakes in Maine continued to show relatively high NO_3^- concentrations. Both the seepage and high-elevation drainage lakes in Maine showed increased DOC concentrations of 10% to 20%, generally by about 0.5 to 1.0 mg/L. The increase in dissolved organic matter would be expected to limit the extent of ANC and pH recovery that would otherwise accompany the observed decreases in SO_4^{2-} concentration. The concentrations of DOC in boreal lakes throughout the northeastern United States, including within ACAD, have increased in recent years. This has been attributed to both decreased S deposition and climate change (Saros 2014). For example, Saros (2014) evaluated the response of algae in two lakes (Jordan Pond, Echo Lake) in ACAD to nutrient (N, P) and DOC addition. Algal growth was co-limited by both N and P in both lakes, and there was no response along a gradient of N input. The added DOC may have stimulated algal growth in Jordan Pond, suggesting that the added DOC contributed growth-limiting nutrients.

The NETN monitors water quality in nine parks, including ACAD, MABI, MIMA, MORR, ROVA, SAGA, SARA, SAIR, and WEFA. Data are summarized at <https://irma.nps.gov/App/Portal/Home>. Sampling is conducted annually from May through October at multiple sites in each park (Gawley et al. 2014). Surface water ANC is measured at all sites to evaluate potential sensitivity to acidification from atmospheric deposition of S and N. Although most states do not have numerical criteria for ANC in their water quality standards, ANC values greater than 100 $\mu\text{eq/L}$ are considered well-buffered, while values less than zero typify acidic waters (Stoddard et al. 2003). Recent network reports summarized data from 2013, placing them in the context of data from 2006-2013.

In ACAD lakes, ponds, and streams, ANC measurements in 2013 followed the typical seasonal pattern of values being lower in spring as compared with summer. This was likely due to the contribution of episodic acidification from spring snowmelt and runoff. Nearly all ANC measurements in ACAD from 2006-2013 were < 100 $\mu\text{eq/L}$, suggesting some sensitivity to acidification. However, in 2013, several lakes and ponds, including Echo Lake (106 $\mu\text{eq/L}$), recorded the highest ANC values measured since the start of NETN monitoring. Seawall Pond had the lowest ANC among the lakes monitored during the period 2006-2013, with several measurements below 20 $\mu\text{eq/L}$. Seawall Pond was an obvious high outlier for most of the water quality parameters measured, perhaps due to its small volume, shallow depth, encircling wetlands, and close (within 60 meters) proximity to the ocean (Gawley and Wiggin 2014). Although upwind acidifying emissions have been reduced since the 1990 CAA, there has been little corresponding change in the ANC and pH of surface waters in ACAD (Bank et al. 2006).

In MABI, one pond and one stream were sampled; all ANC measurements from 2006 to 2013 were higher than 800 µeq/L (Gawley and Roy 2014a). Three streams were sampled in MIMA; all ANC measurements were >200 µeq/L (Gawley and Roy 2014h). Five streams were sampled in MORR; most had ANCs > 400 µeq/L, and all had ANC >100 µeq/L (Gawley and Roy 2014b). Six park streams were sampled in ROVA; all ANC measurements were > 1000 µeq/L (Gawley and Roy 2014c). One pond and two streams were sampled in SAGA; all ANC measurements were > 500 µeq/L (Gawley and Roy 2014d). Two stream sites were sampled in SAIR; most measurements were > 1000 µeq/L, and all were > 400 µeq/L (Gawley and Roy 2014e). Four park streams were sampled in SARA; all measurements were >1000 µeq/L (Gawley and Roy 2014f). One pond, Weir Pond, was sampled at WEFA; all measurements were > 300 µeq/L (Gawley and Roy 2014g). None of these waters show evidence of acid-sensitivity.

Future Projections and Critical Loads for Acidification

Assessments from long-term monitoring sites indicate that the ongoing chemical recovery of Adirondack lakes has slowed since 2000. This observation agrees with model projections, which suggested that continued recovery of water chemistry will likely require additional emissions reductions, beyond those required by regulations in effect as of about 2003. Model projections by Sullivan et al. (2006a) suggested that the ongoing chemical recovery of the more acid-sensitive Adirondack lakes would come to an end over approximately the subsequent decade. Unless air pollution emissions are further reduced, this recovery will likely be followed by a lengthy period of renewed acidification of lakes that were recently recovering from past acidification. This is because recent levels of acidic deposition, even though they are much lower than during the 1970s and 1980s, have nevertheless been high enough that base cations continue to be removed from soils by the SO_4^{2-} and NO_3^- contributed by atmospheric deposition. Two different ecosystem models (MAGIC and PnET-BGC) estimated that the number of Adirondack lakes having an ANC value below 50 µeq/L will increase in the future, even with assumed modest continued reductions in emissions of S (a further 11% decrease) and N (about 20% decrease) below levels that occurred during 2001. However, model estimates suggested that substantial additional future reductions in S and N deposition would result in biological improvements in many of the most severely degraded lakes. Such projected improvements would include the return of one to two species of fish to the lakes, improved conditions for brook trout (*Salvelinus fontinalis*), and an increase by one or two in the number of species of zooplankton. Nevertheless, even if S and N emissions are reduced to half or less of their recent values, the most acid-impacted Adirondack lakes are unlikely to increase to ANC values high enough whereby episodic acidification to ANC below 0 during snowmelt is no longer a threat. Similar responses might be expected for acid-sensitive surface waters in ACAD. However, model simulations have not been conducted for surface waters in that park.

One unfortunate aspect of the response of watersheds to acidic deposition is that the watershed's ability to neutralize acids from acidic deposition changes over time. As more atmospheric acidity is neutralized and more S and N are stored in the soil, the watershed's ability to neutralize and store newly deposited acids is progressively reduced. This has a significant effect on the capacity of watersheds to recover from acidification. In short, the longer emissions controls have been, or will

be, delayed, the less effective those emissions controls will be with respect to recovery of soil and water from acidification.

The Conference of the New England Governors and Eastern Canadian Provinces (NEG/ECP) sponsored development of steady-state CLs for protection of forest soils and lakes against acidification in the NETN region and in eastern Canada (Ouimet et al. 2001, Dupont et al. 2005, Miller 2006, Ouimet et al. 2006). Dupont et al. (2005) used the SSWC steady state aquatic critical loads (CL) model to calculate CLs of acidity and associated exceedances for lakes in the northeastern United States and eastern Canada. Atmospheric acid loads were assessed based on atmospheric deposition of both S and N, using a critical limit of pH=6 to protect aquatic biota. This pH level approximately corresponds with ANC = 40 $\mu\text{eq/L}$ in this region (Small and Sutton 1986). Lakes having lowest calculated CLs included many in southern Vermont, eastern and northern Maine, northern New Hampshire, and Cape Cod. Lakes in Connecticut and parts of Massachusetts exhibited relatively high CLs. Exceedances of CLs, based on estimated acidic deposition in 2002, were highest in central and coastal Massachusetts, southern Vermont, much of Maine, and portions of New Hampshire. Eastern Maine and southern Vermont were notable “hot spots” where ambient S+N deposition exceeded CLs by more than 10 $\text{meq/m}^2/\text{yr}$ (Dupont et al. 2005).

Temporal Variability in Water Chemistry

Episodic acidification of streamwater by NO_3^- has been documented in ACAD by Kahl et al. (1992). Stream pH values in the park have declined during episodes to values as low as 4.7 (Kahl et al. 1992, Heath et al. 1993). The mechanisms that produce acidic episodes can include dilution of base cations and flushing of NO_3^- , SO_4^{2-} , Cl^- , and/or organic acids from forest soils to drainage water (Sullivan et al. 1986, Kahl et al. 1992, Wigington et al. 1996, Wigington 1999, Lawrence 2002). Acidic deposition can contribute to episodic acidification of surface water by supplying N which can produce pulses of NO_3^- during high flow periods, contributing hydrologically mobile SO_4^{2-} through dry deposition, and by lowering baseline pH and ANC, so that episodes are sufficient to produce short-term biologically harmful conditions (Stoddard et al. 2003).

Decreases in pH with increases in flow have been well documented in the NETN region. The most severe acidification of surface waters in this network generally occurs during spring snowmelt (Charles 1991). Stoddard et al. (2003) found that on average, spring ANC values in New England, the Adirondacks, and the Northern Appalachian Plateau were about 30 $\mu\text{eq/L}$ lower than summer values during the period 1990 to 2000 (Figure 1). This implies that lakes and streams in these regions would need to recover to chronic Gran ANC values above about 30 $\mu\text{eq/L}$ before they could be expected to not experience acidic episodes (Stoddard et al. 2003). However, this estimate of 30 $\mu\text{eq/L}$ is certain to be low because the comparison was made with non-episodic sampling in spring.

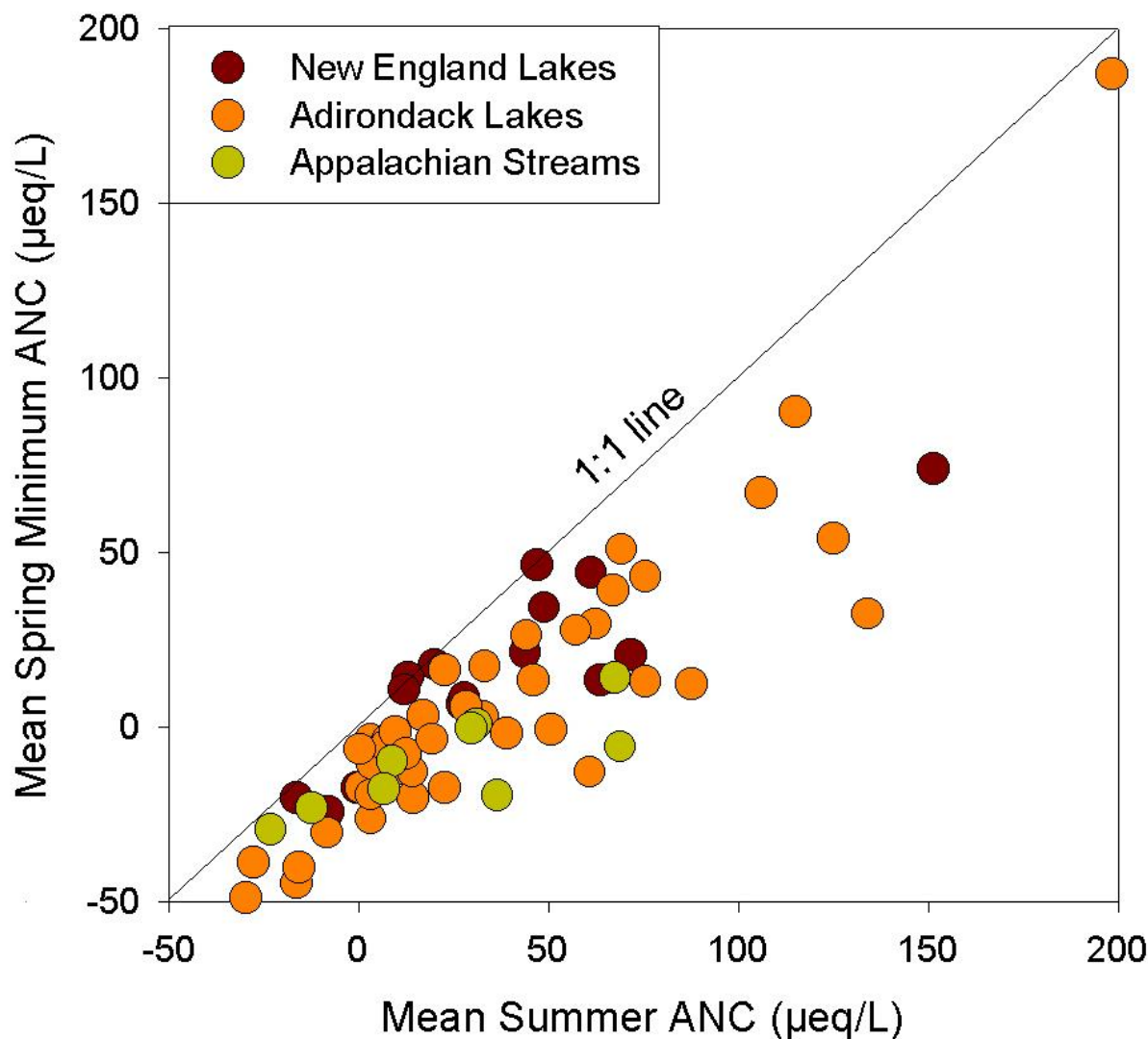


Figure 1. Relationship between mean summer and spring ANC values at LTM sites in New England, the Adirondacks, and Northern Appalachian Plateau. (Source: Stoddard et al. 2003)

The transient nature of high flows and remote location of many sensitive waters makes episodic acidification difficult to measure in the NETN. Therefore, assessments have generally estimated the number of lakes and streams prone to episodic acidification by combining episode information from a few sites with base flow values of ANC determined in large surveys or modeling studies (Eshleman et al. 1995, Bulger et al. 2000, Driscoll et al. 2001). Inclusion of episodically acidified water bodies in regional assessments substantially increases estimates of the extent of surface water acidification.

There is evidence of episodic acidification of headwater streams in ACAD to $\text{pH} < 5.0$, likely in part a result of marine salt and fog deposition to thin, acidic soils (Heath et al. 1992). Salt inputs at ACAD can occur due to marine aerosol deposition and/or road salt application. Vaux et al. (2008) reported higher salt concentrations at sites below roads, as compared with above roads, in six watersheds in ACAD.

A study by Heath et al. (1992) attributed episodic acidification of low-order streams in ACAD primarily to wet and dry atmospheric deposition of sea salts and streamwater dilution, rather than acidic deposition of S and N. Short-term pH depressions of up to 2 pH units and decreases in ANC of up to 130 $\mu\text{eq/L}$ were noted. This appears to have been the first documented example of the “neutral salt” acidification effect in North America. This effect requires acidic soils and inputs of NaCl or $\text{Mg}(\text{Cl})_2$ that facilitate ion exchange in soil solution of Na^+ or Mg^{2+} for H^+ . Coastal fog that is deposited in ACAD is often very acidic, with pH values around 3.0. Storm events and resulting acid episodes occur in ACAD most often in the spring and fall (Bank et al. 2006).

Effects on Aquatic Biota

Aquatic biota in the NETN have been affected by acidification at virtually all levels of the food web. Some species, and some life stages within species, are more sensitive than others. Decreases in ANC and pH and increases in Al_i concentration contribute to declines in species richness and abundance of zooplankton, macroinvertebrates, and fish (Schindler et al. 1985, Keller and Gunn 1995, Nierzwicki-Bauer et al. 2010). Although some species are favored by increased acidity, the overall species pool decreases as surface water acidity increases.

Acidification of surface waters in ACAD from atmospheric pollutant deposition, and the accompanying mobilization of toxic substances such as Al and Hg, can result in an inhospitable stream environment for many species of biota. Bank et al. (2006) reported that the northern dusky salamander (*Desmognathus fuscus fuscus*) has been declining dramatically in ACAD streams. It is possible that this decline is partly due to the effects of toxic Al disrupting the blood Na balance in the salamander and interfering with larval respiration and development (Bank et al. 2006).

Results from the EPA’s Episodic Response Project (ERP) demonstrated that episodic acidification can have long-term adverse effects on fish populations. Streams with suitable chemistry during low flow, but low pH and high Al_i levels during high flow, had substantially lower numbers and biomass of brook trout than were found in non-acidic streams (Wigington et al. 1996). Chemical measurements made during the ERP under high flow correlated with fish community status. In general, reduced trout abundance occurred in ERP streams having median high flow pH < 5.0 and Al_i > 100 to 200 $\mu\text{g/L}$. Acid -sensitive fish species were absent from streams with median high flow pH < 5.2 and Al_i > 100 $\mu\text{g/L}$.

Inorganic Al was the single best predictor of fish mortality in ERP bioassays (van Sickle et al. 1996) and has been identified as an important toxic factor in other bioassays and field studies (Mount et al. 1988, Ingersoll et al. 1990, Rosseland et al. 1990). More recently, Baldigo et al. (2007) found that mortality of brook trout young of the year in the NETN region occurred at Al_i concentrations as low as 54 $\mu\text{g/L}$ (2 μM). The relationships between pH and Al_i or ANC and Al_i vary among streams (Wigington et al. 1996). High Al_i concentrations during episodes are probably the dominant cause of adverse effects on fish during episodic acidity events.

Nutrient Nitrogen Enrichment

The network rankings developed by Sullivan et al. (2011a) in a coarse screening assessment of nutrient N Pollutant Exposure, Ecosystem Sensitivity to nutrient N enrichment, and Park Protection yielded an overall nutrient N enrichment Summary Risk ranking for the NETN that was in the second lowest quintile among all networks. The overall level of concern for nutrient N enrichment effects on I&M parks within the NETN was judged by Sullivan et al. (2011a) to be Low compared to other I&M park networks, based on the coarse-scale available data. Although rankings provide an indication of risk, park-specific data, particularly regarding nutrient-enrichment sensitivity, are needed to fully evaluate risk from nutrient N addition.

Nutrient N Pollutant Exposure rankings for the individual parks in the NETN were variable, from Moderate (middle quintile) for four of the parks to Very High (top quintile) for five parks (Table 3). In contrast, the Ecosystem Sensitivity to nutrient N enrichment rankings were lower (Low to Very Low) for all parks except BOHA, SAIR, and WEFA, which were ranked in the middle quintile for this theme (Table 3). The Ecosystem Sensitivity to nutrient N enrichment rankings of the I&M parks were relatively low because the I&M parks in this network, compared with parks in other networks, have limited coverage of vegetation types (i.e., grassland, arctic herbaceous, alpine, wetland, arid and semi-arid) that are expected to be especially sensitive to nutrient enrichment effects from N deposition, and perhaps also in part because the lakes in most parks within the NETN are generally not at high elevation. High-elevation lakes might be N-limited and more prone than lakes at lower elevation to N- enrichment. High-elevation lakes may therefore be potentially more susceptible to eutrophication in response to atmospheric N input as compared with lower elevation lakes.

Table 3. Estimated park rankings¹ according to risk of nutrient enrichment impacts on sensitive receptors. (Source: Sullivan et al. 2011a)

Park Name	Park Code	Estimated Nutrient N Pollutant Exposure	Estimated Ecosystem Sensitivity to Nutrient N Enrichment
Acadia	ACAD	Moderate	Low
Boston Harbor Islands	BOHA	Very High	Moderate
Home of Franklin D. Roosevelt	HOFR	Very High	Low
Marsh-Billings-Rockefeller	MABI	Moderate	Very Low
Minute Man	MIMA	High	Low
Morristown	MORR	Very High	Very Low
Saint-Gaudens	SAGA	Moderate	Very Low
Saratoga	SARA	Moderate	Low
Saugus Iron Works	SAIR	High	Moderate
Vanderbilt Mansion	VAMA	Very High	Very Low
Weir Farm	WEFA	Very High	Moderate

¹ Relative park rankings are designated according to quintile ranking, among all I&M Parks, from the lowest quintile (Very Low risk) to the highest quintile (Very High risk).

As discussed in the sections that follow, available site-specific data suggest that the potential for nutrient enrichment impacts on terrestrial, and to a lesser extent aquatic, resources in ACAD and

elsewhere within the NETN may be greater than is indicated by the broad regional analysis reported by Sullivan et al. (2011a).

Elvir et al. (2006) investigated the effects of increased N deposition on photosynthesis and foliar nutrient content of sugar maple, American beech, and red spruce at the Bear Brook watershed in Maine. The Bear Brook study is a paired-watershed deposition study in which one watershed was treated beginning in 1989 with bimonthly ammonium sulfate additions at 25.2 kg N/ha/yr. The other watershed served as a reference. Sugar maple, American beech, and red spruce in the treated watershed had higher foliar N concentrations than reference trees (Elvir et al. 2006). American beech and red spruce displayed significantly lower foliar concentrations of Ca, Mg, and Zn. Sugar maple did not show decreases in these nutrients, and was the only species to have significantly higher photosynthetic rates in the treated watershed as compared with the reference. This result suggested that nutrient imbalances in American beech and red spruce may have offset any potential photosynthetic benefits of higher N concentrations (Elvir et al. 2006).

Nitrogen fertilization study plots were established within two adjacent stands at the Harvard Forest research site in Massachusetts. One stand consisted of even-aged red pine planted in 1926. The other was a mixed-deciduous stand that regenerated after a clear-cut in about 1950 and was composed mainly of black and red oak (*Quercus velutina* and *Q. rubra*), followed by black birch (*Betula lenta*), red maple, and American beech (Magill et al. 1997). Each stand contained four plots, three of which received partial applications of fertilizer each month from May to September during year 1 (1988) followed by consistent N treatments applied as NH_4NO_3 each successive year as follows: control, low N (applications of 50 kg N/ha/year), high N (applications of 150 kg N/ha/year) and N+S (applications of 50 kg N/ha/year and 74 kg S/ha/yr; Magill et al. 1997, Magill et al. 2000). After six years of fertilization, hardwood trees had higher wood production and up to 25% higher foliar N concentrations, while pine trees displayed decreased wood production and 67% higher foliar N concentrations (Magill et al. 1997). Nitrate leaching was insignificant from the hardwood plots, but increased continually over the six years in the pine stand, where approximately 85-99% of N was retained and 53% was estimated to be held in the long-term soil pool (Magill et al. 1997). After nine years, N retention efficiency was 97-100% in the control and low N plots, and 96% and 85% in the high N hardwood and pine plots, respectively (Magill et al. 2000). After 15 years, plots that had been continually fertilized with N in both the red pine and the mixed deciduous stands displayed 41% lower soil respiration relative to reference plots (Bowden et al. 2004). Carbon dioxide production in root-free, high N soil from both stands was also significantly lower than in soil in control plots, suggesting that reduced microbial activity contributed to lower soil respiration rates after N addition. Additional effects included decreased forest productivity and higher tree mortality in both forest types (Bowden et al. 2004), decreased needle longevity and about 30-60% lower photosynthetic capacity in conifers (Bauer et al. 2004). Changes were observed in understory plant species assemblages, stem density, biomass, and nutrient content (Rainey et al. 1999).

A study by Sievering et al. (2000) in central Maine investigated how atmospheric N deposition affects forest carbon storage. Net N uptake by the spruce-fir forest canopy was in the range of 1 to 5 kg N/ha/yr, while recycled root N uptake was between 10 and 30 kg N/ha/yr. Because N-availability

is usually limiting for photosynthesis, higher N uptake by the canopy can stimulate enhanced C storage in these and other eastern conifer forests (Sievering et al. 2000).

Nitrogen deposition increases the potential for nutrient enrichment to temperate forest ecosystems in the NETN region because the growth of trees in these systems is often N-limited (Vitousek and Howarth 1991). Atmospheric deposition of N has decreased C:N ratios in soils, and contributed to an increase in net nitrification and associated production of acidity in soils. Nitrogen availability in excess of ecological demand (N saturation) has become common and widespread, as evidenced by elevated NO_3^- concentrations in surface waters during the growing season. In some areas, particularly in high-elevation terrestrial ecosystems which have moved towards a condition of N saturation, high levels of N deposition have caused elevated levels of NO_3^- in drainage waters (Aber et al. 1989, Stoddard 1994, Aber et al. 1998). Elevated NO_3^- leaching contributes to depletion of base cations from forest soils, causing adverse effects on sensitive tree species and acidification of drainage waters in base-poor soils (Aber et al. 1989, Stoddard 1994, Aber et al. 1998). Forests considered sensitive to N deposition include high-elevation red spruce and sugar maple forests which receive high rates of N deposition, and where effects of N deposition on root allocation or late-season growth may exacerbate other stresses from acidic deposition and harsh climate.

The degree of N retention within forested catchments varies with tree species. In a stable isotope tracer (N^{15}) study of four tree species in the Catskill Mountains, Templer et al. (2005) found that N retention is significantly lower in sugar maple stands than in stands of American beech, eastern hemlock, or red oak. However, under experimentally increased N deposition, red oak stands showed a high potential for N leaching, possibly to levels as high as in sugar maple plots (Templer et al. 2005).

The typical immediate response to NH_y ($\text{NH}_3 + \text{NH}_4^+$) addition is enhanced plant growth (Krupa 2003), as long as the NH_y concentrations are below toxic levels. Increased tree growth in response to atmospheric N deposition was documented across the NETN region by Thomas (2010). Such enhanced growth generally occurs mainly above ground (Dueck et al. 1991). This can cause changes in the shoot-to-root N ratio after NH_y uptake. These changes can be detrimental to the plant because they can decrease resistance to environmental stresses such as drought. Addition of NH_y is believed to decrease resistance to drought stress for at least two reasons. First, the demand for C to accompany the assimilated NH_y increases CO_2 uptake through plant leaves and therefore stomata opening and water loss. Second, because shoot growth is more enhanced than root growth by the addition of NH_y , the water supply from the roots can become insufficient to support transpirational water loss during periods of drought (Fangmeier et al. 1994, Krupa 2003). Deposition of NH_y is also believed to reduce frost hardiness of plants (Dueck et al. 1990). This is likely because the addition of NH_y prolongs the growth phase of the plants during autumn and delays winter hardiness. This can cause detrimental effects if the first frost occurs early in the autumn period (Cape et al. 1991). Plants also appear to be more susceptible to fungal infection under high N status or changed nutrient balance such as an increase in the ratio of N to K^+ (Ylimartimo 1991, Krupa 2003).

Thomas et al. (2010) analyzed Forest Inventory Analysis (FIA) data in the NETN region to determine tree growth enhancement across a gradient of atmospheric N deposition from about 3 to 11

kg N/ha/yr. Some tree species showed increased growth across the N input gradient (yellow poplar [*Liriodendron tulipifera*], black cherry [*Prunus serotina*], white ash]). Some showed highest growth at intermediate levels of N deposition (quaking aspen [*Populus tremuloides*] and scarlet oak [*Quercus coccinea*]). Red pine exhibited growth decline across the gradient of increasing N deposition (Thomas et al. 2010). Thus, N deposition at ambient levels can have both positive and negative effects on tree growth, depending on species and deposition level.

Two of the primary indicators of N enrichment in forested watersheds are the leaching of NO_3^- in soil drainage waters and the export of NO_3^- in stream water, especially during the growing season (Stoddard 1994). The concentration of NO_3^- in surface water provides an indication of the extent to which atmospherically deposited N leaches from the terrestrial ecosystem. Some N fertilization experiments suggest that increasing N deposition drives an increase in production of dissolved organic nitrogen (DON) in soil (e.g., Seely and Lajtha 1997, McDowell et al. 2004), but there is little evidence that elevated N deposition increases the export and loss of DON from terrestrial ecosystems. Essentially all of the increase in N export across gradients of N deposition occurs as an increase in NO_3^- rather than DON export. The latter is typically less than 2 kg N/ha/yr from most northeastern forested watersheds (Campbell et al. 2000, Goodale et al. 2000, Lovett et al. 2000, Aber et al. 2003).

In order to understand the effects of added N to forest ecosystems, it is helpful to examine the results of experimental N additions. Experimental N additions to forest ecosystems have elicited positive growth responses in some, but certainly not all, organisms (Emmett 1999, Elvir et al. 2003, DeWalle et al. 2006, Högberg et al. 2006). Forest growth enhancement, to the extent that it occurs, can potentially exacerbate other nutrient deficiencies, such as Ca, Mg, or potassium (K), thereby causing problems with forest health. Multiple long-term experiments have demonstrated transient growth increases followed by increased mortality, especially at higher rates of fertilization (Elvir et al. 2003, Magill et al. 2004, McNulty et al. 2005, Högberg et al. 2006).

Fertilization experiments at Mount Ascutney, VT suggested that N saturation may lead to the replacement of slow-growing spruce-fir forest stands by fast-growing deciduous forests that cycle N more rapidly (McNulty et al. 1996, 2005). At the Bear Brook watershed acidification study site, basal area increment of sugar maple was enhanced 13% to 104% by addition of 25 kg N/ha/yr as $(\text{NH}_4)_2\text{SO}_4$, whereas red spruce was not significantly affected (Elvir et al. 2003). Chronic fertilization combined with drought led to significant mortality in a 70-year old red pine stand at Harvard Forest, Massachusetts (Magill et al. 2004). As red pine died, striped maple (*Acer pensylvanicum*), black cherry, and black birch increased their contributions to annual litterfall production. Parallel fertilization of a 50-year-old red-oak/red maple stand largely stimulated productivity, although the drought in 1995 induced significant mortality in small red maple trees. Fine root biomass was slightly, but not significantly, lower in highly fertilized stands relative to controls in both red pine and oak/maple ecosystems (Magill et al. 2004). Decreased production of fine roots may predispose N-fertilized plants to be more sensitive to intermittent drought, as well as to nutrient depletion exacerbated by acidic deposition.

In a high-elevation red spruce-balsam fir (*Abies balsamea*) forest in the northeastern United States, N fertilization over 14 years led to a decrease in live basal area (LBA) with increasing N additions. In control plots, LBA increased by 9% over the course of the study, while LBA decreased by 18% and 40% in plots treated, respectively, with 15.7 kg N/ha/yr and 31.4 kg N/ha/yr (McNulty et al. 2005). At the Harvard Forest Long Term Ecological Research (LTER) site, at chronic N addition levels of 50 and 150 kg/ha/yr for 15 years, Magill et al. (2004) found 31% and 54% decreases, respectively, in red pine growth.

There is little information available regarding the effects of N deposition on herbaceous plants within northern hardwood forests in the NETN. However, Hurd et al. (1998) reported the results of experimental studies that added N at two- and four-times ambient N deposition at several sites in the Adirondack Mountains. Herbaceous plant coverage decreased after three years of fertilization, largely in response to shading caused by enhanced growth of ferns.

Baron et al. (2011) estimated that 16% of 1,290 Adirondack lakes and 34% of 4,361 New England lakes represented in the Eastern Lakes Survey were likely N-limited, based on having dissolved inorganic N (DIN):total P (TP) ratio (by weight) less than 4 (Table 4). In an analysis of data collected during the mid- to late 1990s from lakes and streams throughout the northeastern United States, Aber et al. (2003) suggested that nearly all N deposition is retained or denitrified in northeastern watersheds that receive less than about 8 to 10 kg N/ha/yr. An analysis of N deposition to forest land in the northeastern United States by Ollinger et al. (1993) suggested that approximately 36% of the forests in the region received 8 kg N/ha/yr or more and may therefore be susceptible to elevated NO_3^- leaching (Driscoll et al. 2003).

Table 4. Proportion of lakes in three nutrient limitation classes (N limited, P limited, and N and P co-limited) based on ratios of dissolved inorganic nitrogen (DIN; $\text{NO}_3^- + \text{NH}_4^+$) to total phosphorus (TP). Data are from the Eastern Lake Survey (Linthurst et al. 1986), conducted in the fall of 1984. (Source: Baron et al. 2011)

Region	Number of Lakes ¹	Number of N Limited Lakes (%) ²	Number of P Limited Lakes (%) ³	Number of Lakes with Co-limited N:P (%) ⁴
Adirondacks	1,290	208 (16%)	777 (60%)	305 (23%)
New England	4,361	1,470 (34%)	1,363 (31%)	1,529 (35%)
Poconos/Catskills	1,506	607 (40%)	559 (37%)	341 (22%)

¹ The Eastern Lake Survey was a stratified random sample of lakes; estimates of the number of lakes in each region are based on the target population sizes.

² Lakes with DIN:TP ratios (by weight) less than four were characterized as N limited, based on the work of Morris and Lewis (1988).

³ Lakes with DIN:TP ratios (by weight) greater than 12 were characterized as P limited, based on the work of Morris and Lewis (1988).

⁴ Lakes with DIN:TP ratios (by weight) between 4 and 12 could not be assigned to nutrient limitation class, and are characterized as either co-limited or limited by something other than N or P.

Most lakes and ponds in and near ACAD are nutrient poor (Kahl et al. 2000). Total P is positively correlated with chlorophyll concentration in Maine lakes and also within the subset that occurs in ACAD, indicating that P is generally limiting to lakes (Vaux et al. 2008). Although eutrophication is a concern in many Maine lakes (Nieratko 1992), there is little evidence that this is occurring to any appreciable extent in ACAD (Vaux et al. 2008).

Aber et al. (2003) found that surface water NO_3^- concentrations exceeded $1 \mu\text{eq/L}$ in watersheds receiving about 9 to 13 kg N/ha/yr of atmospheric N deposition (Figure 2). The lakes and streams found to have high NO_3^- concentration were those receiving N deposition above this range, but responses were variable among those receiving high N deposition. Above this range, mean NO_3^- export increased linearly with increasing deposition at a rate of $0.85 \text{ kg NO}_3\text{-N/ha/yr}$ for every 1 kg N/ha/yr increase in deposition, although there was considerable variability in N retention among watersheds at higher rates of deposition (Figure 3; Aber et al. 2003).

Nutrient enrichment experiments were conducted by Levine et al. (1997) to test the limitation status of Lake Champlain, located between New York and Vermont. Response of phytoplankton to nutrient additions varied by month and season. In spring, phytoplankton abundance did not respond to any nutrient additions. Separate additions of P and N in June each increased phytoplankton biomass in the lake, but these same additions in May had no effect. In July and September, N addition had a minor impact and P addition did not affect phytoplankton biomass at all. Additions of N along with P in the summer and fall always resulted in higher phytoplankton biomass than additions of P alone.

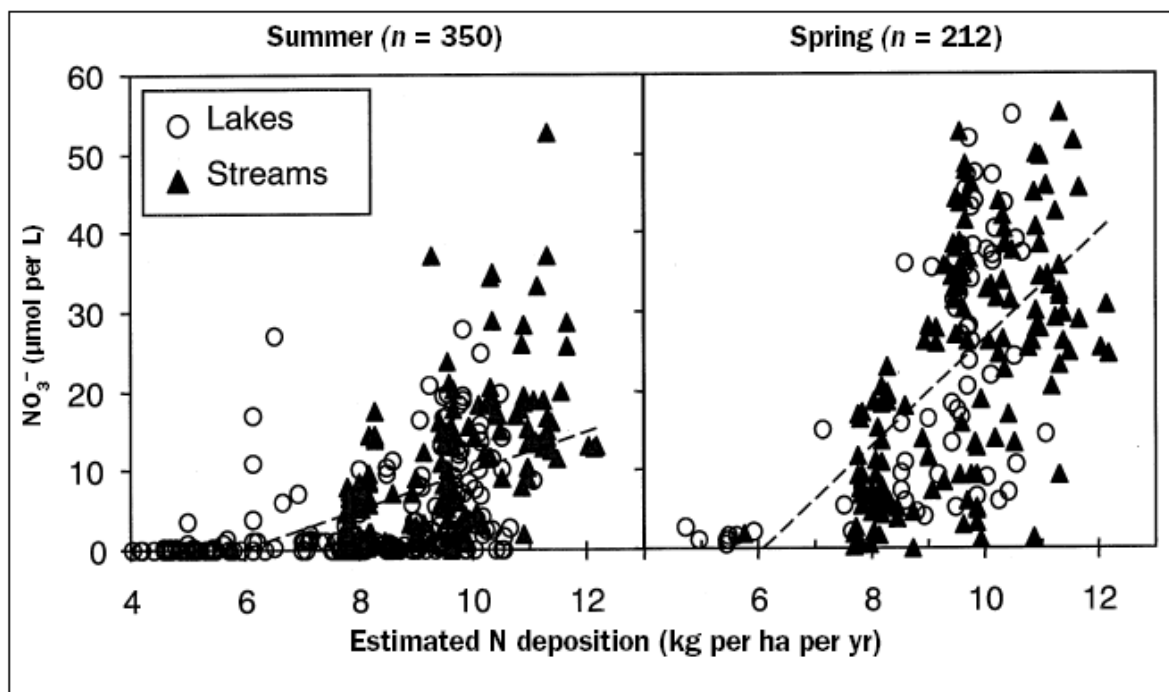


Figure 2. Surface water nitrate (NO_3^-) concentrations as a function of nitrogen deposition at the base of each watershed in summer and spring. Nitrogen deposition to the whole watershed may be 2 to 6 kg/ha/yr greater than at the base. (Source: Aber et al. 2003)

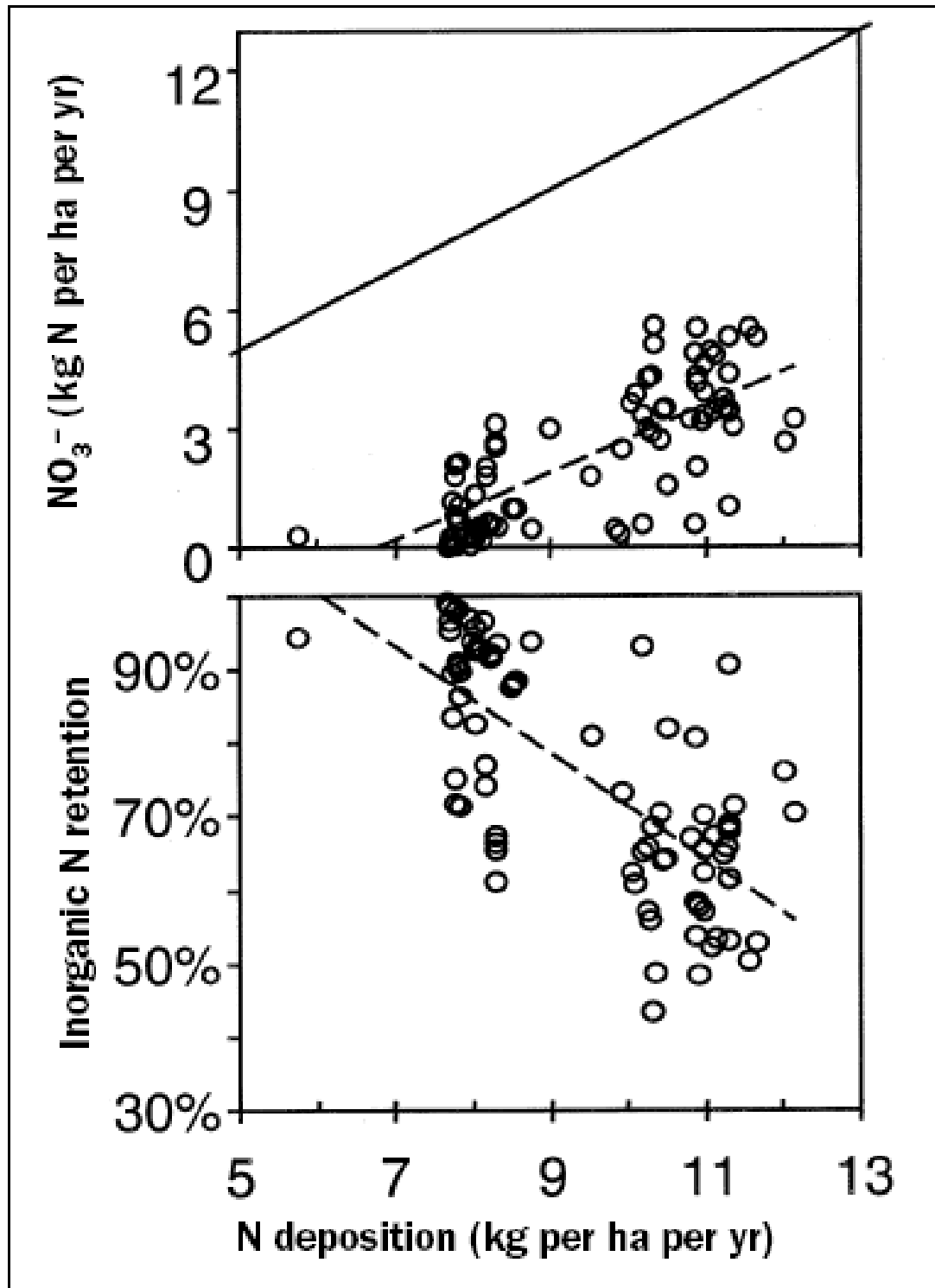


Figure 3. (a) Nitrogen export in stream water as a function of N deposition at the base of sampled watersheds. Nitrogen export is represented by the equation $\text{NBexp} = 0.85 \text{ Ndep} - 5.8$; $r^2 = 0.56$; $p < 0.001$. (b) Watershed N retention decreases as N deposition increases at the base of the watersheds ($\text{N retention} = -0.07 \text{ N deposition} + 1.44$; $r^2 = 0.50$). (Source: Aber et al. 2003)

There was evidence for spatial and temporal variability in the influence of P on phytoplankton growth, with both P sufficiency and P deficiency commonly evident throughout the lake. The authors determined that summer phytoplankton growth is primarily P limited, but that N is also an important factor that influences the biomass and community composition of phytoplankton in Lake Champlain (Levine et al. 1997).

In most upland forested areas in the northeastern United States, most N received in atmospheric deposition is retained in soil (Nadelhoffer et al. 1999). Several different data compilations indicated that 80% to 100% of N deposition is retained or denitrified within terrestrial ecosystems that receive less than about 10 kg N/ha/yr (Dise and Wright 1995, Sullivan 2000, MacDonald et al. 2002, Aber et al. 2003, Kristensen et al. 2004). Nitrogen-related adverse effects on aquatic life occur despite retention of most deposited N within the terrestrial environment (Driscoll et al. 2003). For example, although 70% to 88% of atmospheric N deposition was retained in Catskill Mountains watersheds, fish populations could not be sustained because high NO_3^- concentrations in stream water during high flows caused the concentrations of Al_i to exceed the toxicity threshold (Lawrence et al. 1999).

Researchers at the University of Maine and USGS conducted a paired watershed study in ACAD to investigate the effects on N retention of the fires in 1947 that burned about a third of the park. The unburned study watershed exported 10 to 20 times more inorganic N than the burned watershed (Nelson et al. 2007) and retention of inorganic N was 96% in the burned watershed compared with 72% in the unburned watershed (Campbell et al. 2004).

At the whole-ecosystem experiment at the Bear Brook watershed, experimental $(\text{NH}_4)_2\text{SO}_4$ addition over a period of 10 years had minimal effect on stream detritus processing (Chadwick and Huryn 2003). The N additions had no significant effect on stream macroinvertebrate secondary production or varying production by functional feeding groups. The researchers concluded that climate-related variables such as flow duration and litter inputs controlled secondary production when N was not limiting (Chadwick and Huryn 2005). Data from eight years of experimental $(\text{NH}_4)_2\text{SO}_4$ additions at the Bear Brook watershed were analyzed by Parker et al. (2001) in order to investigate the effects of elevated N deposition on forest-floor C/N ratios. Results showed that the experimentally acidified watershed had lower C and N pools as compared with the reference watershed, and forest-floor C/N ratios in the treated watershed were significantly reduced from 30.6 to 23.4 (Parker et al. 2001).

Peat-forming bog ecosystems are among the most sensitive transitional ecosystems to the effects of N deposition. In the conterminous United States, peat-forming bogs are most common in areas that were glaciated, including in portions of the NETN (U.S. EPA, 1993). Other types of wetlands occur across broad areas of the United States. It is not clear whether current levels of N deposition cause extensive eutrophication to these ecosystems.

Some wetland ecosystems, especially ombrotrophic bogs and nutrient-poor fens, receive most of their nutrient inputs from atmospheric deposition. These wetlands have been shown to experience changes in plant species composition in response to high levels of atmospheric N deposition. The risk of species composition change is important, in part because wetland ecosystems often contain a relatively large number of rare plant species.

For two ombrotrophic bogs in Vermont and Massachusetts, model estimates of extinction risk for the northern pitcher plant (*Sarracenia purpurea*) were calculated by Gotelli and Ellison (2002) under varying N deposition scenarios. Modeled extinction risk within the next 100 years was relatively low under precipitation N concentrations equivalent to those at the time of the study (0.391 to 0.477 mg N/L/yr). However, model estimates showed increased extinction risk under elevated N deposition scenarios (Gotelli and Ellison 2002).

Data are not available with which to evaluate the extent to which wetlands in the NETN have been affected by nutrient enrichment from N deposition. Wetlands are widely distributed within the NETN region, including some areas that receive moderate levels of N deposition. Wetlands comprise about 11% of park lands in ACAD, classified as follows (Calhoun et al. 1994):

Marine	37.5%
Palustrine	31.6%
Estuarine	20.0%
Lacustrine	10.7%
Riverine	<1%

Ambient levels of N deposition may or may not be sufficiently high to cause species shifts in wetland plants in ACAD and elsewhere within the NETN. If such shifts do occur, they most likely occur in palustrine and lacustrine wetlands such as bogs and poor fens that normally receive most of their nutrients from atmospheric inputs.

Nutrient loading to coastal ecosystems in the NETN is an important concern (Nielsen and Kahl 2007), and the issue of nutrient loading to coastal waters was identified by ACAD as a high-priority resource management issue for the park (Kahl et al. 2000). Many estuaries in this network region have become increasingly eutrophic, largely in response to N loading from nonpoint pollution sources (Howes et al. 1996, Nixon 1996, Kinney and Roman 1998). Data from ACAD are important in this context because there have been few studies of coastal New England streams that have little human influence other than atmospheric input to serve as benchmarks (Nielsen and Kahl 2007).

Watershed export of N and P to coastal waters around Mt. Desert Island in Maine, in and around ACAD, may be greatly affected by land use history and human influence. Total N and total P exported by watersheds entirely within ACAD were significantly lower than exports by watersheds that were partly or completely outside of the park (Nielsen and Kahl 2007).

Pardo et al. (2011b) compiled data on empirical CL for protecting sensitive resources in Level I ecoregions across the conterminous United States against nutrient enrichment effects caused by atmospheric N deposition. Available data on empirical CL of nutrient-N in the NETN suggested that the lower end of estimates of the CL for resource protection was largely between about 3 and 8 kg N/ha/yr (Table 5). This level of loading considered to be potentially problematic pertained to protection of mycorrhizal fungi, lichens, and forest vegetation and also prevention of NO₃⁻ leaching into drainage water. Pardo et al. (2011b) estimated that ambient N deposition was consistently higher than these CL values. Thus, these empirical CL data suggest the possibility of widespread exceedance of nutrient-N CL within parks in the NETN.

Table 5. Empirical critical loads for nitrogen in the NETN, by ecoregion and receptor from Pardo et al. (2011b). Ambient N deposition reported by Pardo et al. (2011b) is compared to the lowest critical load for a receptor to identify potential exceedance, indicated by graying. A critical load exceedance suggests that the receptor is at increased risk for harmful effects.

NPS Unit	Ecoregion	N Deposition (kg N/ha/yr)	Critical Load (kg N/ha/yr)				
			Mycorrhizal Fungi	Lichen	Herbaceous Plant	Forest	Nitrate Leaching
Acadia NP	Eastern Temperate Forests	5.2	5 - 12	4 - 8	17.5	3 - 8	8
Boston Harbor Islands NRA	Eastern Temperate Forests	8.9	5 - 12	4 - 8	17.5	3 - 8	8
Eleanor Roosevelt NHS	Eastern Temperate Forests	11.3	5 - 12	4 - 8	17.5	3 - 8	8
Home of Franklin D. Roosevelt NHS	Eastern Temperate Forests	11.3	5 - 12	4 - 8	17.5	3 - 8	8
Marsh-Billings-Rockefeller NHP	Northern Forests	9.4	5 - 7	4 - 6	7 - 21	3 - 26	8
Minute Man NHP	Eastern Temperate Forests	14.2	5 - 12	4 - 8	17.5	3 - 8	8
Morristown NHP	Eastern Temperate Forests	16.2	5 - 12	4 - 8	17.5	3 - 8	8
Morristown NHP	Northern Forests	16.2	5 - 7	4 - 6	7 - 21	3 - 26	8
Saint-Gaudens NHS	Northern Forests	8.2	5 - 7	4 - 6	7 - 21	3 - 26	8
Saugus Iron Works NHS	Eastern Temperate Forests	8.9	5 - 12	4 - 8	17.5	3 - 8	8
Saratoga NHP	Eastern Temperate Forests	12.0	5 - 12	4 - 8	17.5	3 - 8	8
Vanderbilt Mansion NHS	Eastern Temperate Forests	11.3	5 - 12	4 - 8	17.5	3 - 8	8
Weir Farm NHS	Eastern Temperate Forests	16.8	5 - 12	4 - 8	17.5	3 - 8	8

Ellis et al. (2013) estimated the CL for nutrient-N deposition to protect the most sensitive ecosystem receptors in 45 national parks. The lowest terrestrial CL of N was generally estimated for protection of lichens (Geiser et al. 2010). Changes to lichen communities may signal the beginning of other changes to the ecosystem that might affect structure and function (Pardo et al. 2011a). Ellis et al. (2013) estimated the N CL for ACAD in the range of 3-8 kg N/ha/yr for protection of hardwood forests.

Ozone Injury to Vegetation

Ozone levels are high throughout much of the Northeast. Ozone pollution can harm human health, reduce plant growth, and cause visible injury to foliage. Two of the NETN parks, MORR and WEFA, are in counties designated by EPA as nonattainment for the O₃ standard because of their high O₃ levels. The O₃-sensitive plant species that are known or thought to occur within the I&M parks found in the NETN are listed in Table 6. Those considered to be bioindicators exhibit distinctive symptoms when injured by O₃ (e.g., dark stipple) and are designated by an asterisk in the table. Each park within the network contained at least 10 O₃-sensitive and/or bioindicator species. ACAD contained 17 sensitive species, 11 of which are recognized as bioindicators.

Table 6. Ozone-sensitive and bioindicator plant species known or thought to occur in the I&M parks of the NETN. (Data Source: E. Porter, National Park Service, pers. comm., August 30, 2012); lists are periodically updated at <https://irma.nps.gov/NPSpecies/Report>.

Species	Common Name	Park										
		ACAD	BOHA	HOFR	MABI	MIMA	MORR	SAGA	SAIR	SARA	VAMA	WEFA
<i>Aesculus octandra</i>	Yellow buckeye						x					
<i>Ailanthus altissima</i> *	Tree-of-heaven		x	x		x	x		x		x	x
<i>Alnus rugosa</i> *	Speckled alder					x	x	x		x	x	x
<i>Apios americana</i> *	Groundnut	x	x	x	x	x	x	x	x	x	x	x
<i>Apocynum androsaemifolium</i> *	Spreading dogbane	x	x		x	x	x	x		x		x
<i>Apocynum cannabinum</i>	Dogbane, Indian hemp		x		x	x	x	x		x		x
<i>Asclepias exaltata</i> *	Tall milkweed					x	x	x				
<i>Asclepias incarnata</i>	Swamp milkweed		x		x	x		x	x	x		
<i>Asclepias syriaca</i> *	Common milkweed	x	x	x	x	x	x	x	x	x	x	x
<i>Aster acuminatus</i> *	Whorled aster				x	x		x				x
<i>Aster macrophyllus</i> *	Big-leaf aster	x			x	x	x	x				
<i>Cercis canadensis</i> *	Redbud						x					
<i>Clematis virginiana</i>	Virgin's bower			x	x			x		x		x
<i>Corylus americana</i> *	American hazelnut			x		x	x			x	x	
<i>Eupatorium rugosum</i> *	White snakeroot		x	x	x		x			x	x	
<i>Fraxinus americana</i> *	White ash	x	x	x	x	x	x	x		x	x	x
<i>Fraxinus pennsylvanica</i>	Green ash	x	x	x	x	x	x		x	x	x	x
<i>Gaylussacia baccata</i> *	Black huckleberry	x	x			x	x			x	x	x
<i>Liquidambar styraciflua</i>	Sweetgum					x						
<i>Liriodendron tulipifera</i> *	Yellow-poplar		x	x		x	x				x	x
<i>Lyonia ligustrina</i> *	Maleberry					x	x			x		x
<i>Parthenocissus quinquefolia</i>	Virginia creeper	x	x	x	x	x	x		x	x	x	x
<i>Philadelphus coronarius</i>	Sweet mock orange		x				x			x	x	
<i>Pinus banksiana</i>	Jack pine	x										
<i>Pinus rigida</i>	Pitch pine	x	x	x		x	x			x	x	

* Bioindicator species

Table 6 (continued). Ozone-sensitive and bioindicator plant species known or thought to occur in the I&M parks of the NETN. (Data Source: E. Porter, National Park Service, pers. comm., August 30, 2012); lists are periodically updated at <https://irma.nps.gov/NPSpecies/Report>).

Species	Common Name	Park										
		ACAD	BOHA	HOFR	MABI	MIMA	MORR	SAGA	SAIR	SARA	VAMA	WEFA
<i>Pinus virginiana</i>	Virginia pine						x					
<i>Platanus occidentalis</i> *	American sycamore		x	x	x	x	x	x		x	x	
<i>Populus tremuloides</i> *	Quaking aspen	x	x		x	x	x	x		x		x
<i>Prunus serotina</i> *	Black cherry	x	x	x	x	x	x	x	x	x	x	x
<i>Prunus virginiana</i>	Choke cherry	x	x	x	x	x	x	x	x	x	x	
<i>Rhus copallina</i>	Winged sumac						x					
<i>Robinia pseudoacacia</i>	Black locust		x	x	x	x	x		x	x	x	x
<i>Rubus allegheniensis</i> *	Allegheny blackberry	x	x	x	x	x	x	x		x	x	x
<i>Rubus cuneifolius</i>	Sand blackberry							x				
<i>Rudbeckia laciniata</i> *	Cutleaf coneflower			x						x		
<i>Sambucus canadensis</i> *	American elder	x	x	x	x	x	x			x	x	x
<i>Sambucus racemosa</i> *	Red elderberry				x							
<i>Sassafras albidum</i>	Sassafras		x	x		x	x			x	x	x
<i>Solidago altissima</i>	Goldenrod		x			x	x			x		
<i>Spartina alterniflora</i>	Smooth cordgrass	x	x									
<i>Symphoricarpos albus</i> *	Common snowberry	x	x								x	
<i>Vitis labrusca</i> *	Northern fox grape		x			x	x		x		x	x

* Bioindicator species

From 1983 to 2005, O₃ levels at the McFarland Hill monitoring site in ACAD exceeded the O₃ standard of 0.08 ppm (based on an 8-hr average) in effect at that time (Vaux et al. 2008) one or more times each year. In 2004, EPA designated Hancock County, ME, where ACAD is located, as a non-attainment area for O₃. Between 1995 and 2005, the mean annual number of 8- hour O₃ exceedances was 4.6 at the Cadillac Mountain monitoring site (Vaux et al. 2008). Because of various air quality improvement programs stemming from the CAA and its amendments, O₃ levels decreased at ACAD and elsewhere in the Northeast. Hancock County was redesignated as a Maintenance Area in 2007. Since that time, O₃ levels have continued to decrease (NPS-ARD 2013) and Hancock County is more recently in compliance with even the more stringent 2008 O₃ standard of 0.075 ppm.

In the past, polluted air masses from the industrialized northeastern corridor of the United States created elevated O₃ exposure conditions in the NETN during the growing season that could potentially damage sensitive plant species. Forests in ACAD have experienced periodic episodes of O₃ above 80 ppbv throughout the summer months, and foliar symptoms were observed in several plant species in the 1990s (Bartholomay et al. 1997). Ozone has been shown to reduce photosynthesis in white pine, even in the absence of visible foliar injury (Spence et al. 1990, Kozlowski et al. 1991). Bartholomay et al. (1997) used dendroclimatic techniques to investigate the relationship between O₃

exposure in ACAD and white pine (*Pinus strobus*) growth rates. Regression analysis indicated that O₃ exposure explained more variation in tree growth than did climate. This may have been because elevated O₃ concentrations can disrupt photosynthetic rates and reduce carbohydrate availability (Bartholomay et al. 1997).

The W126 (a measure of cumulative O₃ exposure that preferentially weights higher concentrations) and SUM06 (a measure of cumulative exposure that includes only hourly concentrations over 60 ppb O₃) exposure indices calculated by NPS staff for NETN parks are given in Table 7, along with Kohut's (2007) O₃ risk ranking. The NPS and Kohut ranking systems differ. The NPS ranking system (NPS 2010) is a quick assessment of O₃ condition that ranks O₃ exposure levels according to injury thresholds from the literature (Heck and Cowling 1997), using a 5-year average of either the W126 or SUM06 index. Both metrics are calculated over a 3-month period. The W126 was classified as Moderate exposure at values between 7 and 13 ppm-hr, as defined by NPS (2010). Values higher than 13 ppm-hr were classified as High exposure, and values lower than 7 ppm-hr were classified as Low exposure. The SUM06 was classified as Moderate at values between 8 and 15 ppm-hr. Higher and lower values were classified as High and Low, respectively, as defined by NPS (2010).

Kohut's approach constitutes a more rigorous assessment of potential risk to plants. It considers both O₃ exposure and environmental conditions (soil moisture). Kohut also used injury thresholds from the literature, but evaluated a different O₃ metric (after Lefohn et al. 1997), the W126 over a 5-month period in conjunction with the N100 (number of hours over 100 ppb O₃). The rationale for the N100 statistic is that higher O₃ concentrations are most likely to cause plant injury. Kohut examined five individual years of O₃ exposure and soil moisture data and considered the effects of low soil moisture on O₃ uptake each year when assigning risk. Soil moisture is important because dry conditions induce stomatal closure in plants, which has the effect of limiting O₃ uptake and injury. In areas where low soil moisture levels correspond with high O₃ exposure, uptake and injury are limited by stomatal closure even when exposures are relatively high. Ozone condition, as rated by NPS, ranges from Low to Moderate in all of the NETN parks except MORR, which was ranked High based on the SUM06 index. Kohut's evaluation of risk to plants ranged from Low in three parks to High in four parks (HOFR, MORR, VAMA, and WEFA). ACAD was ranked Moderate by Kohut.

Table 7. Ozone assessment results for I&M parks in the NETN based on estimated average 3-month W126 and SUM06 ozone exposure indices for the period 2005-2009 and Kohut's (2007) ozone risk ranking for the period 1995-1999¹.

Park Name	Park Code	W126		Sum06		Kohut O ₃ Risk Ranking
		Value (ppm-hr)	Ranking	Value (ppm-hr)	Ranking	
Acadia	ACAD	4.40	Low	4.78	Low	Moderate
Boston Harbor Islands	BOHA	8.74	Moderate	10.79	Moderate	Moderate
Home of Franklin D. Roosevelt	HOFR	11.48	Moderate	12.31	Moderate	High
Marsh-Billings-Rockefeller	MABI	4.93	Low	5.42	Low	Low
Minute Man	MIMA	8.57	Moderate	10.57	Moderate	Moderate
Morristown	MORR	12.37	Moderate	15.74	High	High
Saint-Gaudens	SAGA	5.04	Low	5.59	Low	Low
Saratoga	SARA	6.93	Low	8.03	Moderate	Low
Saugus Iron Works	SAIR	8.41	Moderate	10.34	Moderate	Moderate
Vanderbilt Mansion	VAMA	11.47	Moderate	11.69	Moderate	High
Weir Farm	WEFA	11.52	Moderate	14.14	Moderate	High

¹ Parks are classified into one of three ranks (Low, Moderate, High), based on comparison with other I&M parks.

The results of both ranking systems should be considered when evaluating the potential for O₃ injury to park vegetation. The Kohut approach considered environmental conditions that significantly affect plant response to O₃, but exposures have likely changed since the time of the assessment. The NPS approach considers more recent O₃ conditions, but not environmental conditions.

In the eastern United States, ground level O₃ formation is controlled to a greater extent by NO_x emissions than by VOCs (NRC 1992, Ryerson et al. 2001). Indirect effects of NO_x emissions through ground-level O₃ production include reduction in net photosynthetic capacity (Reich 1987) and associated changes in biomass production and carbon allocation (Laurence et al. 1994). Ozone related decreases in above-ground forest growth in the NETN in the 1990s appeared to be in the range of negligible to 10% per year, based on analyses reported by Chappelka and Samuelson (1998). In recent years, decreases in O₃ exposure levels would be expected to alleviate these previous growth effects.

Extrapolation from seedling-level experiments is complicated and uncertain, but process-level modeling of O₃ effects offers promise for combining physiological effects with stand- and site-level factors. An analysis involving the PnET ecosystem model estimated that O₃ exposure in the northeastern United States reduced annual rates of NPP by 2% to 16% in the 1990s. Variation resulted mainly from differences in O₃ exposure levels, and interactions with other atmospheric pollutants (Ollinger et al. 1997, 2002). This analysis did not reflect more recent conditions, which indicate significantly reduced O₃ levels.

Visibility Degradation

Natural Background and Ambient Visibility Conditions

The CAA set a specific goal for visibility protection in Class I areas: “the prevention of any future, and the remedying of any existing, impairment of visibility in mandatory Class I federal areas which impairment results from manmade air pollution” (42 U.S.C. 7491). In 1999, EPA passed the Regional Haze Rule (RHR), which requires each state to develop a plan to improve visibility in Class I areas, with the goal of returning visibility to natural conditions in 2064. Natural background visibility assumes no human-caused pollution, but varies with natural processes such as windblown dust, fire, volcanic activity and biogenic emissions. Visibility is monitored by the Interagency Monitoring of Protected Visual Environments Network (IMPROVE) and typically reported using the haze index *dv*. Although ACAD is the only Class I park in the NETN, improvements in visibility required by the RHR at ACAD and other Class I areas in the Northeast are expected to benefit air quality and visibility in the other parks as well.

Natural background and ambient haze levels have been estimated by IMPROVE for one park in the NETN, ACAD. Data are also available from Cape Cod National Seashore that are considered to be representative of visibility conditions for two of the other parks, BOHA and SAIR. A monitoring site is considered by IMPROVE to be representative of an area if it is within 60 mi (100 km) and 425 ft (130 m) in elevation of that area. Relative to all I&M parks, ACAD, BOHA, and SAIR had very high natural haze for the 20% clearest natural haze conditions, 20% haziest natural haze conditions, and for the average of all natural haze conditions (Table 8). Natural haze levels tend to be higher in coastal areas in part due to the influence of sea salt aerosols and marine SO_4^{2-} .

Ambient visibility estimates reflect pollution levels and were used to rank conditions at parks in order to provide park managers with information on spatial differences in visibility and air pollution. Rankings range from very low haze (very good visibility) to very high haze (very poor visibility). Only parks with on-site or representative IMPROVE monitors were used in generating the baseline visibility ranking. Table 8 gives the relative park haze rankings on the 20% clearest, 20% haziest, and average days. The relative ranking for measured ambient haze for the period 2004 through 2008 was High for all parks monitored within the NETN for all groups (20% clearest, 20% haziest, and average). Deciview¹ (*dv*) values in all three monitored parks were higher (about 3.5 to 11 *dv*) than natural haze values (Table 8).

¹ The *deciview* visibility metric expresses uniform changes in haziness in terms of common increments across the entire range of visibility conditions, from pristine to extremely hazy conditions. Because each unit change in deciview represents a common change in perception, the deciview scale is like the decibel scale for sound. A one deciview change in haziness is a small but noticeable change in haziness under most circumstances when viewing scenes in Class I areas.

Table 8. Estimated natural haze and measured ambient haze in I&M parks in the NETN averaged over the period 2004 through 20081. The IMPROVE site at Cape Cod National Seashore (CACA1) is considered representative of conditions at BOHA and SAIR.

Park Name	Park Code	Site ID	Estimated Natural Haze					
			20% Clearest Days	20% Haziest Days	Average Days			
			dv	dv	dv			
Acadia	ACAD	ACAD1	4.66	12.43	7.88			
Boston Harbor Islands ²	BOHA	CACO1	5.95	13.20	9.08			
Home of Franklin D. Roosevelt	HOFR	No Site						
Marsh-Billings-Rockefeller	MABI	No Site						
Minute Man	MIMA	No Site						
Morristown	MORR	No Site						
Saint-Gaudens	SAGA	No Site						
Saratoga	SARA	No Site						
Saugus Iron Works	SAIR	CACO1	5.95	13.20	9.08			
Vanderbilt Mansion	VAMA	No Site						
Weir Farm	WEFA	No Site						
Park Name	Park Code	Site ID	Measured Ambient Haze (For Years 2004 through 2008)					
			20% Clearest Days		20% Haziest Days		Average Days	
			dv	Ranking	dv	Ranking	dv	Ranking
Acadia	ACAD	ACAD1	8.06	High	22.06	High	14.01	High
Boston Harbor Islands ²	BOHA	CACO1	11.18	High	24.27	High	16.64	High
Home of Franklin D. Roosevelt	HOFR	No Site						
Marsh-Billings-Rockefeller	MABI	No Site						
Minute Man	MIMA	No Site						
Morristown	MORR	No Site						
Saint-Gaudens	SAGA	No Site						
Saratoga	SARA	No Site						
Saugus Iron Works	SAIR	CACO1	11.18	High	24.27	High	16.64	High
Vanderbilt Mansion	VAMA	No Site						
Weir Farm	WEFA	No Site						

¹ Parks are classified into one of five ranks (Very Low, Low, Moderate, High, Very High).

² Data are from a representative IMPROVE site. A monitoring site is considered by IMPROVE to be representative of an area if it is within 60 mi (100 km) and 425 ft (130 m) in elevation of that area.

Acadia

20% Clearest Days

Taken: 3:00 PM

Haze = 8 dv

$B_{\text{ext}} = 22 \text{ Mm}^{-1}$

VR = 180 km



20% Haziest Days

Taken: 3:00 PM

Haze = 22 dv

$B_{\text{ext}} = 87 \text{ Mm}^{-1}$

VR = 35 km



Average Days

Taken: 3:00 PM

Haze = 15 dv

$B_{\text{ext}} = 43 \text{ Mm}^{-1}$

VR = 90 km



Figure 4. Three representative photos of the same view in ACAD illustrating the 20% clearest visibility, the 20% haziest visibility, and the annual average visibility. B_{ext} is total particulate light extinction; VR is visual range.

Representative photos of a selected vista in ACAD under three different visibility conditions are shown in Figure 4. Photos were selected to correspond with the clearest 20% of visibility conditions, haziest 20% of visibility conditions, and annual average visibility conditions at that location. This series of photos provides a graphic illustration of the visual effect of these differences in haze level on a representative vista in this park.

IMPROVE data allow estimation of visual range (VR). Data from ACAD indicate that air pollution has reduced average VR in the park from 100 to 50 miles (161 to 80 km). On the haziest days, VR has been reduced from 70 to 20 miles (113 to 24 km). Severe haze episodes occasionally reduce visibility to 6 miles (10 km). At CACO, representative of conditions at BOHA and SAIR, pollution has reduced average VR from 95 to 40 miles (153 to 64 km). On the haziest days, VR has been reduced from 60 to 20 miles (97 to 32 km). Severe haze episodes occasionally reduce visibility to 6 miles (10 km).

Composition of Haze

Various pollutants make up the haze that causes visibility degradation. IMPROVE measures these pollutants and reports them as ammonium sulfate, ammonium nitrate, elemental carbon, coarse mass, organic mass, sea salt, and soil. Sulfates form in the atmosphere largely from SO₂ emissions released by coal-burning power plants, smelters, and other industrial facilities. Nitrates form in the atmosphere from NO_x emissions from combustion sources including vehicles, power plants, industry, and fires. Organic compounds are emitted from a variety of both natural (biogenic) and anthropogenic sources, including agriculture, industry, and fires. Atmospheric sea salt concentrations are higher in coastal areas. Soil can enter the atmosphere through both natural processes and human disturbance.

Sulfate is the most important cause of fine particle pollution and visibility impairment² across the MANE-VU states, including within the NETN region. At the Class I sites in this region, SO₄²⁻ accounts for about one-half to two-thirds of total fine particle mass on the 20% haziest days (Northeast States for Coordinated Air Use Management (NESAUM) 2006). On the 20% clearest days, SO₄²⁻ typically accounts for 40% or more of total fine particle mass. Regional SO₄²⁻ emissions generally control visibility in the MANE-VU region during summer. During winter, visibility depends more on both regional and local S sources and also on local meteorological conditions (including temperature inversions). Organic C is, after SO₄²⁻, the next most important contributor to regional haze within MANE-VU (NESAUM 2006).

The majority of total particulate light extinction (b_{ext}) in ACAD was attributable to SO₄²⁻ and organics (Figure 5). On average, SO₄²⁻ contributed 62.3% of total b_{ext} , and organics contributed 14.4%. On the 20% haziest days, the contribution of SO₄²⁻ increased to 69.2% of b_{ext} , and organics decreased to 12.4%. On the clearest 20% visibility days, the contribution of SO₄²⁻ decreased to 51.3%, and organics increased to 19.6% of b_{ext} . In BOHA and SAIR, SO₄²⁻, NO₃⁻, organics, and sea

² *Visibility impairment* means any humanly perceptible change in visibility (light extinction, visual range, contrast, coloration) from that which would have existed under natural conditions.

salt were the major contributors to b_{ext} . Sea salt extinction was proportionally more important on the 20% clearest days, contributing 16.2% compared to 3.9% on the 20% haziest days in these two parks (Figure 5).

Visibility at ACAD varies with wind direction. Days having clearest visibility tend to be those when air masses derive from the north. Haziest visibility occurs on days when air masses derive from the south and southwest (Vaux et al. 2008).

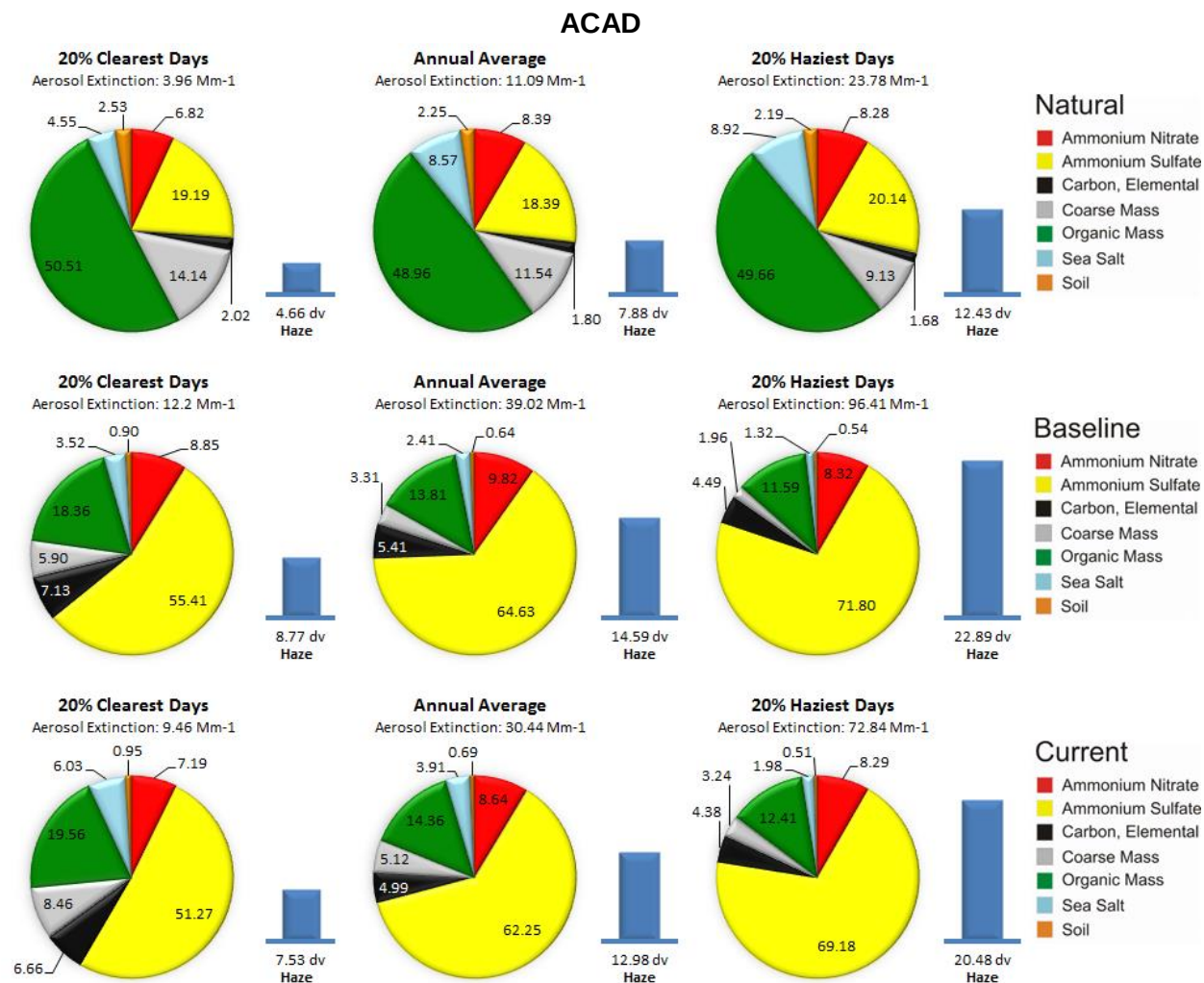


Figure 5a. Estimated natural (pre-industrial), baseline (2000-2004), and current (2006-2010) levels of haze (blue columns) and its composition (pie charts) on the 20% clearest, annual average, and 20% haziest visibility days for ACAD. Data for BOHA and SAIR were taken from a site considered representative (CACO1); there are no data for the years 2000 and 2001. (Data Source: NOPS-ARD)

BOHA and SAIR (data from CACO1)

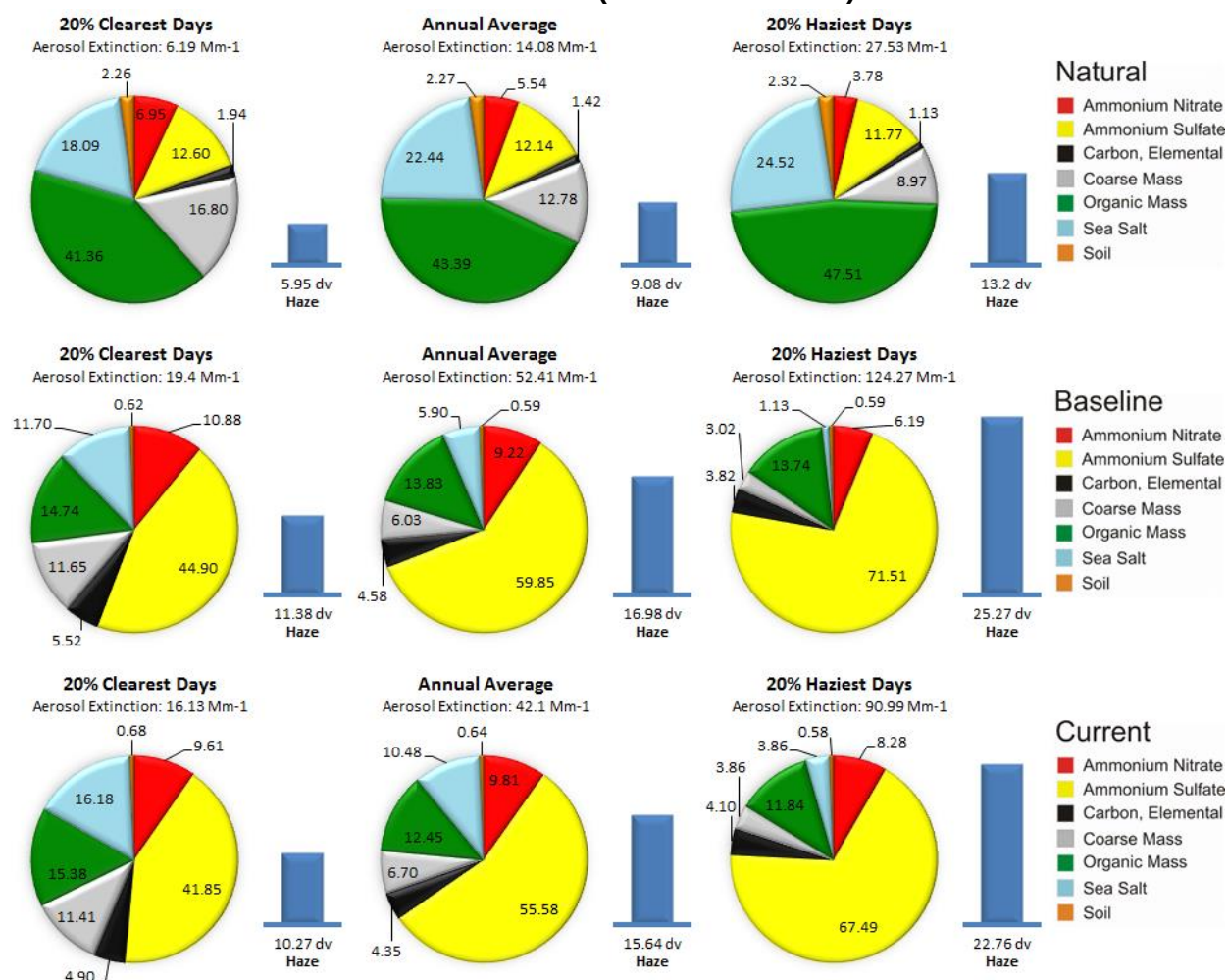


Figure 5b. Estimated natural (pre-industrial), baseline (2000-2004), and current (2006-2010) levels of haze (blue columns) and its composition (pie charts) on the 20% clearest, annual average, and 20% haziest visibility days for BOHA and SAIR. Data for BOHA and SAIR were taken from a site considered representative (CACO1); there are no data for the years 2000 and 2001. (Data Source: NOPS-ARD)

Trends in Visibility

NPS (2010) reported long-term trends in annual dv on the clearest and haziest 20% of days at monitoring sites in 29 national parks. All 27 parks that showed statistically significant ($p \leq 0.05$) trends on the 20% clearest days for the 11-20 year monitoring periods through 2008 exhibited decreases in haze over time. None of the sites showed increasing trends on the clearest days. The steepest declines (-0.18 to -0.20 dv/yr) on the clearest days were reported for Shenandoah National Park (SHEN), ACAD, and Washington DC, with 18-19 years of monitoring data at each of those locations. Ten parks showed statistically significant decreases in dv on the haziest days, with the steepest declines reported for Mount Rainier National Park (MORA; -0.38 dv/yr), SHEN (-0.27 dv/yr), ACAD (-0.26 dv/yr), and Washington DC (-0.25 dv/yr), with 18-19 years of data for each of those locations.

Annual mean haze levels on the haziest days at 47 monitored park locations during the period 2006-2008 ranged from 1.5 to 21 dv higher than the estimated natural condition (NPS 2010). The average difference between measured dv and estimated natural condition was 8.3 dv. Several eastern parks, including ACAD, had annual mean dv on the haziest days that were substantially higher (more than about 10 dv) than estimated natural conditions.

Within the northeastern United States, visibility improvement during recent years has been most pronounced in the western and southern portions of the MANE-VU region, in and near the NETN. The area of greatest visibility improvement has been in proximity to large power plant sources of SO₂ emissions in the Ohio River and Tennessee Valleys (NESCAUM 2006). Between 1996 and 2006, visibility in scenic areas improved on the days that have the haziest visibility at five locations in the United States. One of those locations was the Great Gulf Wilderness in New Hampshire, within the NETN region (U.S. EPA 2008).

Trends in haze on the 20% clearest, average, and 20% haziest days are shown in Figure 6 for ACAD, BOHA, and SAIR. Visibility at ACAD on the 20% clearest, 20% haziest, and average days has been improving throughout the period of monitoring record since 1990 (Figure 6). The steepest declines in ambient haze at ACAD have occurred on the 20% haziest days (~ 8 dv improvement). Progress has also been substantial at this park on the 20% clearest (~ 5 dv) and average days (~ 7 dv). Decreases in ambient haze have also been documented at BOHA and SAIR, mainly since 2005. Improvements in visibility at these parks have been less pronounced than at ACAD.

Development of State Implementation Plans

MANE-VU is using a weight of evidence approach to meeting the requirements of the RHR. Multiple independent methods are used to assess the relative contribution of different emissions sources and source regions to regional haze in the Class I areas within and near the MANE-VU region (NESCAUM 2006). These approaches include application of Eulerian grid-based source models, Lagrangian (air parcel-based) source dispersion models, back trajectory calculations, and analysis of monitoring data.

According to the RHR, states and tribes must establish and meet reasonable progress goals for each federal Class I area to improve visibility on the 20% haziest days and to prevent visibility degradation on the 20% clearest days. The national goal is to return visibility in Class I areas to natural background levels in 2064. States must evaluate progress by 2018 (and every 10 years thereafter) based on a baseline period of 2000 to 2004 (Air Resource Specialists 2007).

A variety of model estimates suggested that emissions from the MANE-VU states account for about 25 to 30% of the fine particulate SO₄²⁻ observed in ACAD and other Class I sites within the MANE-VU region. The Midwest Regional Planning Organization (MWRPO) and Visibility Improvement State and Tribal Association of the Southeast (VISTAS) states each accounted for another approximately 15% of total fine particulate SO₄²⁻ at ACAD and about 25% each at other Class I areas within the MANE-VU region (NESCAUM 2006). Although MANE-VU is focusing primarily on

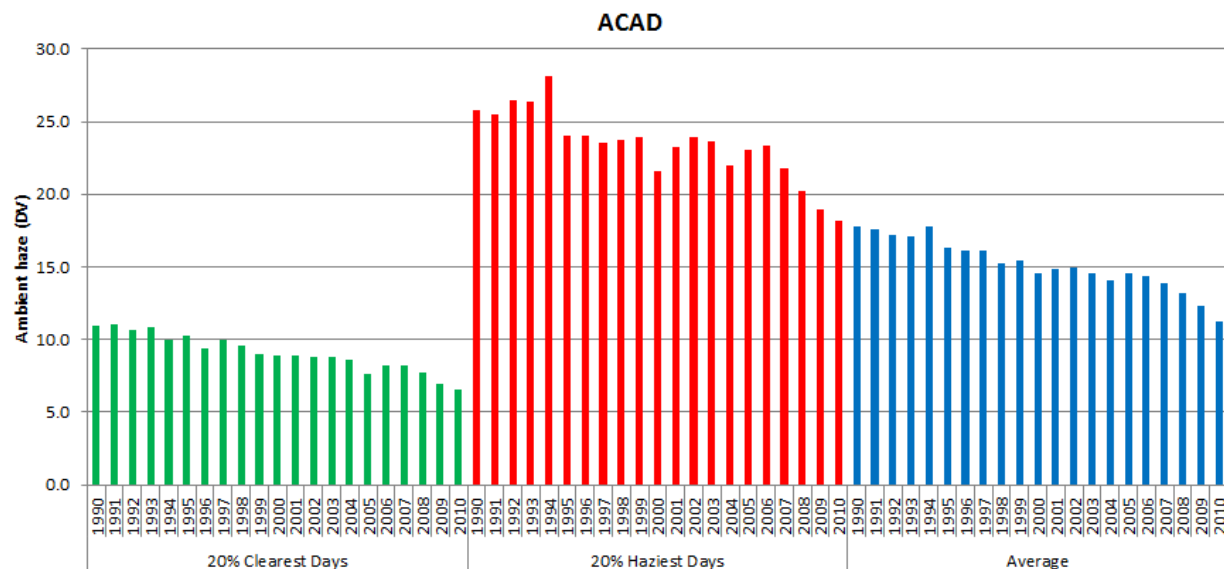


Figure 6a. Trends in ambient haze levels at ACAD, based on IMPROVE measurements on the 20% clearest, 20% haziest, and annual average visibility days over the monitoring period of record. Data for BOHA and SAIR were taken from a nearby site (CACO1). Data Source: http://vista.cira.colostate.edu/improve/Data/IMPROVE/summary_data.htm

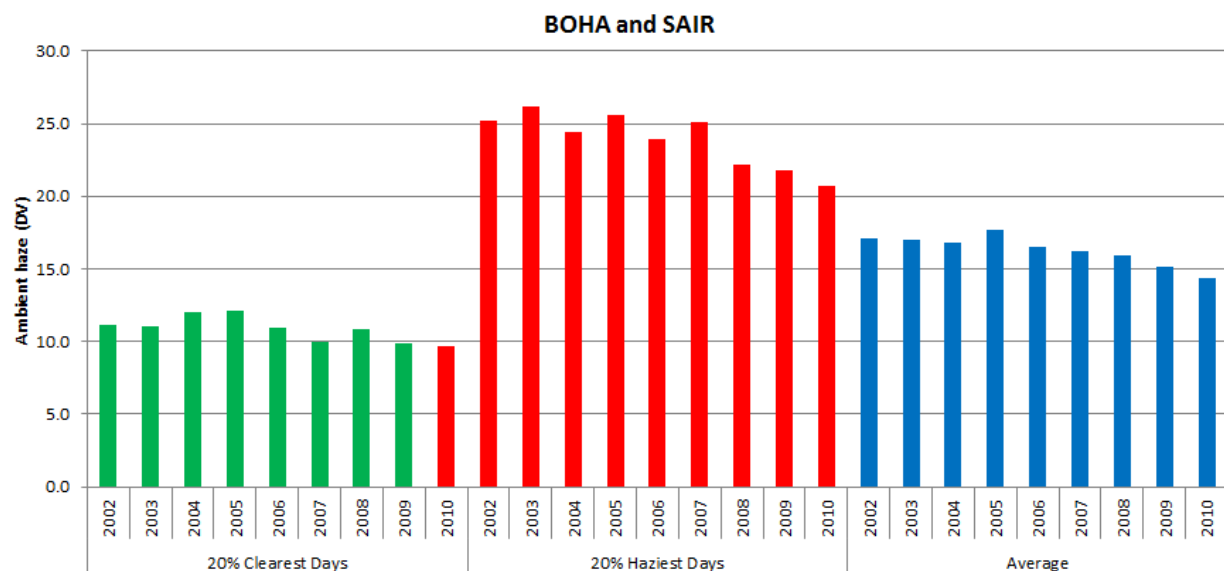


Figure 6b. Trends in ambient haze levels at BOHA and SAIR, based on IMPROVE measurements on the 20% clearest, 20% haziest, and annual average visibility days over the monitoring period of record. Data for BOHA and SAIR were taken from a nearby site (CACO1). Data Source: http://vista.cira.colostate.edu/improve/Data/IMPROVE/summary_data.htm

fine particle SO_4^{2-} abatement in its initial efforts toward compliance with the RHR (NESCAUM 2006), controls on other haze-producing constituents, such as organic C and both urban and mobile sources of NO_x during winter will also be important.

Progress to date in meeting the national visibility goal is illustrated in Figure 7 using a uniform rate of progress glideslope. Improvements in visibility on the 20% haziest days at all of the monitored parks in the NETN have so far been sufficient to comply with the glideslope requirements of the RHR. Additional monitoring will be required to assure continued compliance.

Toxic Airborne Contaminants

Estimates of Hg methylation potential generated by USGS (2015) for watershed boundaries (based on eight-digit hydrologic unit codes [HUCs]) containing national park lands in the northern portion of the NETN region suggested high methylation potential at many parks considered in this analysis (Figure 8). Estimates were highest in and around ACAD, but were also high at SAIR, MIMA, BOHA, and WEFA. This result is likely driven mainly by relatively high concentrations of total organic carbon in surface waters in this network.

Much of the research in the United States that has been conducted on Hg methylation, the influence of SO_4^{2-} on methylation rates, and controls on Hg transport within watersheds has been conducted in two park network areas, one of which is the NETN (cf., Chen et al. 2005, Evers 2005, Evers et al. 2005, Kamman et al. 2005, Driscoll et al. 2007b, Evers et al. 2007, Evers et al. 2008). Myers et al. (2007) proposed a national map showing the relative sensitivity of aquatic ecosystems to Hg contamination. Areas showing the highest estimated sensitivity included portions of the NETN region. ACAD has been the focus of substantial research on Hg biogeochemistry.

The ACAD region lies within a coastal zone of generally low levels of Hg in surface water compared with the rest of Maine (Peckenham et al. 2007). However, measured values of Hg in ACAD were more typical of higher concentrations that are found in northwestern Maine. Thus, Mt. Desert Island represents a hotspot of locally high Hg within the regional context (Vaux et al. 2008). Other parks in the network are located well to the south of this hotspot. Total dissolved Hg was measured in streams across Mt. Desert Island by Peckenham (2007). Highest concentrations were recorded in Squid Cove Brook, Oak Hill Stream, Hodgdon Brook, and Whalesback Brook. Hodgdon Brook is a tributary to Hodgdon Pond, where the highest Hg concentration in a Maine fish was found in 1995 (Vaux et al. 2008).

Research conducted in ACAD suggested that fire may play a role in Hg cycling within forested watersheds in the NETN (Amirbahman et al. 2004). Nelson et al. (2007) investigated the effects of fire on Hg and N dynamics at two watersheds in ACAD. The reference watershed, Hadlock Brook, has been largely undisturbed for the past 300 years and is dominated by red spruce and balsam fir (*Abies balsamea*; Schauffler et al. 2007). In contrast, the Cadillac Brook watershed was the site of a 1947 stand-replacing wildfire that decreased soil organic matter. The burned watershed supports a heterogeneous mix of hardwood forest and softwood stands (Schauffler et al. 2007).

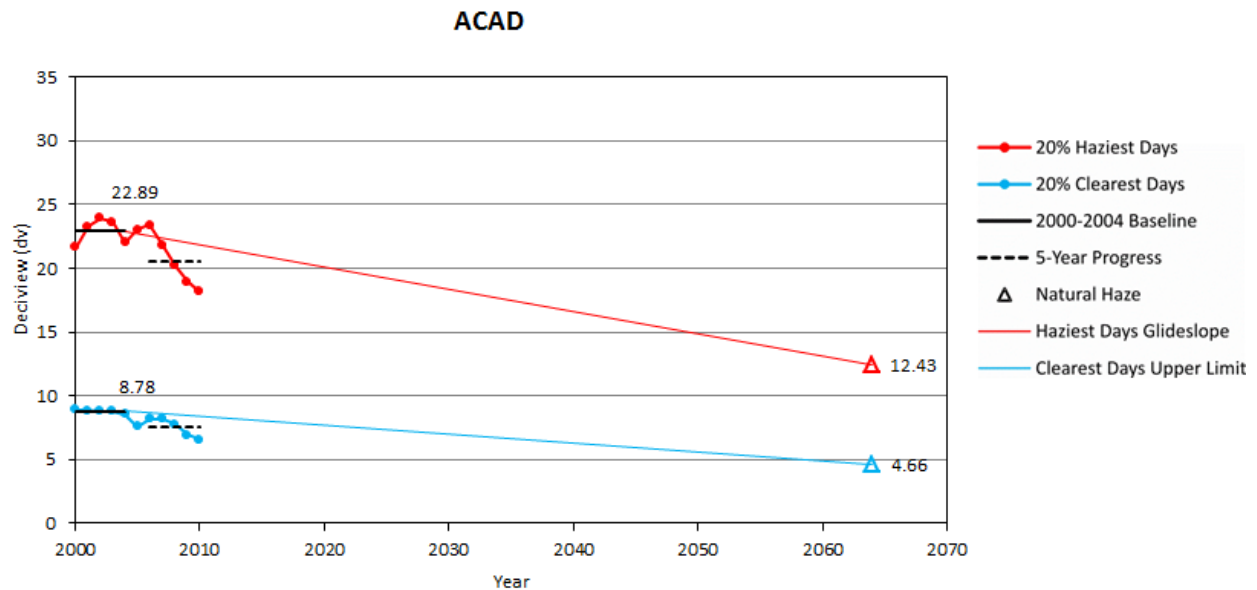


Figure 7a. Glideslopes to achieving natural visibility conditions in 2064 for the 20% haziest (red line) and the 20% clearest (blue line) days in ACAD. In the Regional Haze Rule, the clearest days do not have a uniform rate of progress glideslope; the rule only requires that the clearest days do not get any worse than the baseline period. Also shown are measured values during the period 2000 to 2010. Data for BOHA and SAIR were taken from a representative site (CACO1) and have no data for the years 2000 and 2001. Data Source: http://vista.cira.colostate.edu/improve/Data/IMPROVE/summary_data.htm

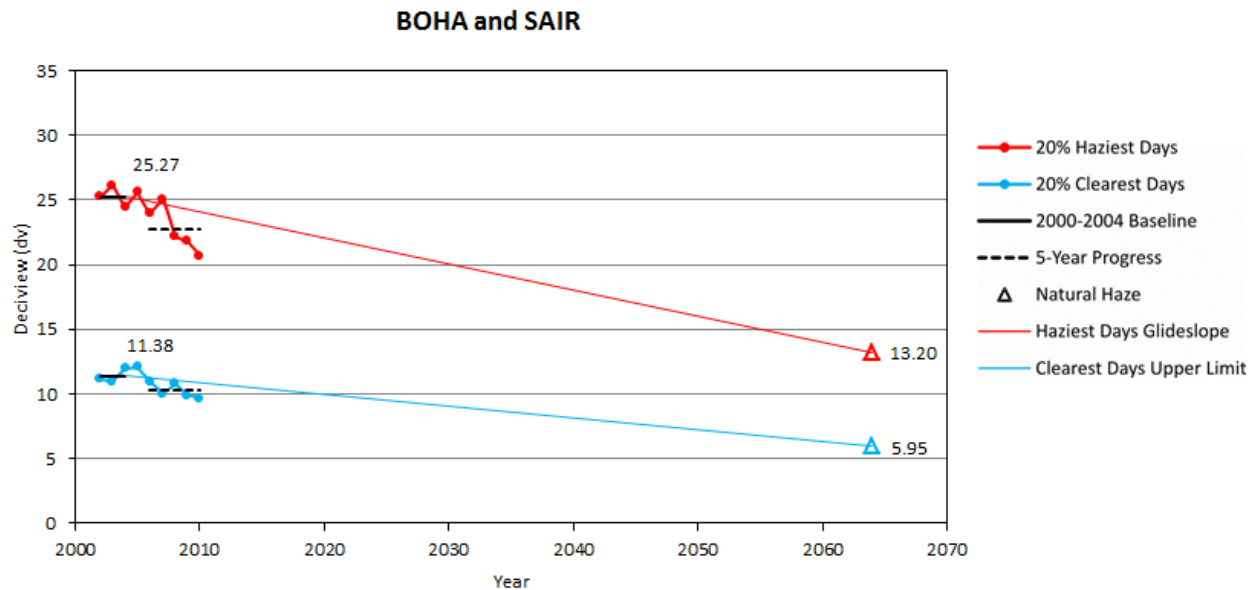
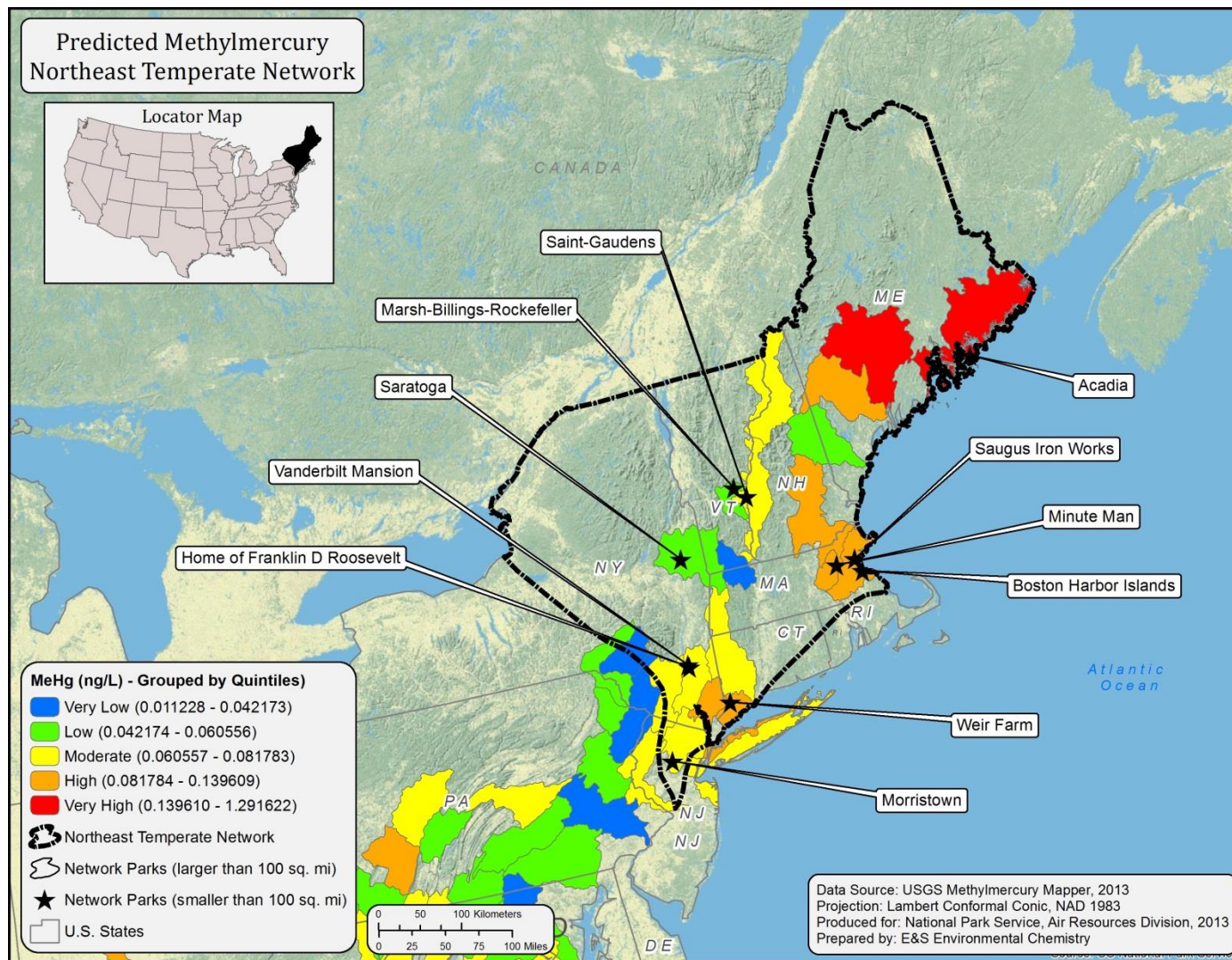


Figure 7b. Glideslopes to achieving natural visibility conditions in 2064 for the 20% haziest (red line) and the 20% clearest (blue line) days in BOHA and SAIR. In the Regional Haze Rule, the clearest days do not have a uniform rate of progress glideslope; the rule only requires that the clearest days do not get any worse than the baseline period. Also shown are measured values during the period 2000 to 2010. Data for BOHA and SAIR were taken from a representative site (CACO1) and have no data for the years 2000 and 2001. Data Source: http://vista.cira.colostate.edu/improve/Data/IMPROVE/summary_data.htm



Map 9. Predicted MeHg concentrations in surface waters by HUCs that contain national parklands in the Northeast Temperate Network. Estimates were generated by USGS (2015). Rankings are based on quintile distributions across all I&M parks having estimates by USGS.

Analysis of streamwater and precipitation samples collected from 1999 to 2000 showed that Hg transport was lower in the watershed recovering from fire damage ($0.4 \mu\text{g}/\text{m}^2/\text{year}$ at Cadillac Brook versus $1.3 \mu\text{g}/\text{m}^2/\text{year}$ at Hadlock Brook). This was also true for dissolved inorganic N (DIN) export, which was $11.5 \text{ eq}/\text{ha}/\text{year}$ from Cadillac Brook and $92.5 \text{ eq}/\text{ha}/\text{year}$ from Hadlock Brook (Nelson et al. 2007). Higher Hg deposition occurred in the Hadlock Brook watershed, where mature conifer stands are more efficient at scavenging Hg from the atmosphere relative to the bare soil, exposed bedrock, and regenerating hardwood vegetation that occurs throughout much of the Cadillac Brook watershed (Nelson et al. 2007).

Peckenham et al. (2007) found that Hg concentrations in streamwater in ACAD were statistically correlated with both the amount of upstream wetland in the watershed and the total DOC in streamwater. Mercury concentrations were highest in lower-elevation portions of streams, likely due to cumulative inputs from wetlands and riparian areas (Peckenham et al. 2007). This study also reported that headwater forested watersheds are more efficient in transporting Hg per unit DOC, which to some extent counterbalances the higher total DOC generated in wetlands.

The NPS sampled surface waters in ACAD to distinguish water quality effects from local organic and metal pollutant sources (in particular, traffic-related emissions) versus long-range atmospheric transport and deposition (Peckenham et al. 2006). Samples were collected across a gradient from immediately below high-use roads to remote areas of the park. Differences between busy roads and remote sources were used to estimate the importance of local sources of pollutants. They did not detect any polynuclear aromatic hydrocarbon (PAH) or volatile organic compounds above $1 \mu\text{g}/\text{L}$. Low concentrations of metals that are known to be associated with motor fuels were detected at all sample locations. The locations of sites showing the highest metal detection occurrences suggested a likely important contribution from vehicles to localized metal loading (Peckenham et al. 2006).

The northern Great Lakes region, and eastward into northern New York, is especially sensitive to Hg bioaccumulation, due in part to relatively high Hg deposition and in particular due to watershed and lake characteristics that enhance Hg transport, methylation, and bioaccumulation (Evers et al. 2011a).

In the northeastern United States, high concentrations of Hg in yellow perch and common loon have been shown to be significantly correlated with water chemistry: total P $< 30 \mu\text{g}/\text{L}$, DOC $> 4 \text{ mg}/\text{L}$, pH < 6.0 , ANC $< 100 \mu\text{eq}/\text{L}$ (Chen et al. 2005, Driscoll et al. 2007b). High Hg concentrations have also been shown to be correlated with several landscape characteristics: wetland abundance, low lake:watershed area ratio, high percent forest cover, and high atmospheric Hg deposition (Roué-LeGall et al. 2005, Driscoll et al. 2007b). These characteristics affect Hg transport, methylation, and trophic transfer (St. Louis et al. 1996, Wiener et al. 2006, Driscoll et al. 2007b, Turnquist et al. 2011).

Webber and Haines (2003) investigated the biological and behavioral effects of MeHg exposure on fish (golden shiner [*Notemigonus crysoleucas*]) collected from Brewer, ME. Results indicated that, although growth rate and mortality did not differ with varying MeHg loads, fish with increased MeHg exposure displayed less efficient predator-avoidance behaviors. Webber and Haines (2003) concluded that Hg exposure at levels found in the NETN region can alter fish predator avoidance

behavior and may increase vulnerability to predation. This has implications regarding food chain transfer of Hg to piscivorous wildlife.

Elevated MeHg accumulation in fish-eating birds in the northeastern United States has been linked to lake acidification (Evers et al. 2007). Methylation is critical to the effects of Hg on aquatic biota; this form of Hg is toxic, bioavailable, and accumulates in top predators to levels of concern for both human health and the environment (Table 9; Evers et al. 2007).

Table 9. Summary statistics of biological data layers for mercury (Hg) concentrations in fish and wildlife ($\mu\text{g per g}$) in the northeastern United States and southeastern Canada. (Source: Evers et al. 2007)

Category/Species	Sample Size	Data Layer Designation	Hg Concentrations		Hg Level of Concern (Tissue Type)	Percentage of Samples with Concentration > Level of Concern
			Mean ± Standard Deviation	Range		
Human health						
Yellow perch ¹	4089	Primary	0.39 ± 0.49	< 0.05-5.24	0.30 (fillet)	50
Largemouth bass ²	934	Secondary	0.54 ± 0.35	<0.05-2.66	0.30 (fillet)	75
Ecological health						
Brook trout	319	Secondary	0.31 ±0.28	<0.05-2.07	0.16 (whole fish)	75
Yellow perch ³	(841) ⁴	Secondary	0.23 ± 0.35	<0.05-3.18	0.16 (whole fish)	48
Common loon ⁵	1546	Primary	1.74 ± 1.20	0.11-14.20	3.0 (blood)	11
Bald eagle	217	Secondary	0.52 ± 0.20	0.08-1.27	1.0 (blood)	6
Mink	126	Secondary	19.50 ± 12.1	2.80-68.50	30.0 (fur)	11
River otter	80	Secondary	20.20 ± 9.30	1.14-37.80	30.0 (fur)	15

Note: All data are in wet weight except for fur, which is on a fresh-weight basis

¹ Fillet Hg in yellow perch is based on individuals with a standardized length of 20 cm.

² Fillet Hg in largemouth bass is based on individuals with a standardized length of 36 cm.

³ Whole-fish Hg in yellow perch is based on individuals with a standardized length of 13 cm. Whole-fish Hg for yellow perch was converted to fillet Hg.

⁴ The sample population of 841 yellow perch examined for whole-fish Hg is included with the 4089 fillets (i.e., the total number of all biotic data layers does not double-count yellow perch).

⁵ Egg Hg for the common loon was converted to the adult blood equivalent

Methylation is correlated with multiple aspects of water acid-base chemistry (Wiener et al. 2006, Driscoll et al. 2007b); methylating bacteria generally require SO_4^{2-} to carry on their metabolic activities, so increased S deposition has been shown to increase rates of Hg methylation in fresh water wetlands (Galloway and Branfireun 2004, ICF International 2006, Jeremiason et al. 2006). Wetlands act as important sources of MeHg to fresh water ecosystems. This is likely due in large part

to two characteristics of wetlands: 1) high availability of DOC, and 2) anaerobic conditions in sediments. Both enhance methylation rates. Mercury binds to organic matter. As a consequence, DOC is an important parameter affecting Hg bioavailability and transport through watersheds (Grigal 2002). The abundance of DOC enhances the transport of MeHg to downstream receiving waters. Many lakes, especially small lakes in the NETN region, have extensive wetlands in their watersheds and contain relatively high concentrations of DOC (cf., Linthurst et al. 1986, Kretser et al. 1989). As a consequence of wetland influences on Hg methylation and transport, the percentage of wetland area within watersheds is commonly correlated with MeHg flux (Grigal 2002).

Accumulation of MeHg in fish-eating birds can result in damage to nervous, excretory, and reproductive systems (Wolfe et al. 1998). Table 10 lists several studies indicating effects related to mercury bioaccumulation in avian eggs and tissues. Reproduction is considered one of the most sensitive endpoints for chronic low-level MeHg exposure of fish-eating birds (Wolfe et al. 1998). Reduced clutch size, increased number of eggs laid outside the nest, eggshell thinning, and increased embryo mortality have all been documented (Wolfe et al. 1998). Kramar et al. (2005) determined that the extent of wetland located in close proximity (less than 150 m) to loon territory was positively correlated with Hg concentrations in loon blood.

The lowest observed adverse effect level (LOAEL) provides a benchmark for quantifying potential injury to wildlife from Hg exposure. For example, the LOAEL for the common loon has been established as 3.0 µg/g of Hg in adult loon blood (Evers et al. 2007, Evers et al. 2008). This level of Hg is associated with reproductive effects such as reduced fledgling success (Burgess and Meyer 2008). However, loon Hg exposure data can include either sex; different ages, locations, and time periods; and can be derived from analysis of blood, tissue or eggs. It has therefore been difficult to standardize the data regarding MeHg availability. Evers et al. (2011b) developed linkages among loon Hg measurements in eggs, blood, and fish prey in the Great Lakes region, including the western portion of the NETN region. Data were normalized into standard loon tissue units. Use of a standard unit of measure that combines multiple tissues of a high profile species and its principal prey items facilitates examination of spatial gradients in pollution effects (Evers et al. 2011b). Based on analysis of over 8,000 male loon units (MLUs), seven biological Hg hotspots were identified in the region. The average MLU concentration across the region was 1.8 µg/g; 82% were above 1 µg/g and 9.8% were above the LOAEL of 3 µg/g. Evers et al. (2011b) examined Hg data in greater detail for four focal regions where rationales for these biological hotspots had been identified. The focal area having the highest average MLU was northern New York (2.38 µg/g; Evers et al. 2011b).

At a given lake, MLUs tend to have higher Hg concentration than female loon units (FLUs) because male loons are typically 21% larger than female loons (Evers et al. 2010) and therefore eat larger fish (Barr 1996). These larger fish tend to have higher Hg concentration (Sandheinrich and Wiener 2011).

The biological Hg hotspot identified in northern New York by Evers et al. (2011b) was characterized by mixed deciduous and coniferous forest (73%) and scrub-shrub wetlands including sphagnum bogs. Lake pH and ANC tend to be low (Yu et al. 2011). The prevalence of wetlands is an important component of Hg methylation, and therefore bioaccumulation, in this area (St. Louis et al. 1994, Kramar et al. 2005). Over 20% of the loon population in this hotspot region has been estimated to be

at potential risk of adverse effects of Hg on reproduction (Evers et al. 2011b). As a consequence, there may be long-term adverse effects on loon populations (Evers et al. 2004, Burgess and Meyer 2008, Evers et al. 2008).

Table 10. Mercury concentrations in avian eggs and tissues and related effects. Source: Wolfe et al. (1998)

Wet (w)					
Tissue	Concen. (ppm)	or Dry (d)	Endpoint	Species	Reference
Liver	1.06	w	No effect	Common tern	Gochfeld (1980)
Liver	22.2	w	Abnormal feather loss in juveniles	Common tern	Gochfeld (1980)
Liver	5	w	Conservative threshold for major toxic effects	Water birds	Zillioux (1993)
Liver	7.2	w	Increased disease and emaciation	Common tern	Spalding (1991)
Liver	9.08	w	Nesting success	Common tern	Finley (1978)
Liver	20.7	w	Hatching success	Common tern	Finley (1978)
Liver	30	w	Neurologic effects	Osprey	Heinz (1974)
Liver	35	w	Death	Common loon	Wiemeyer (1987)
Liver	54.5	w	LD33 ¹	European starling	Finley (1979)
Liver	97.7	w	Death	Gannet	
Liver	103.6	w	LD33	European starling	Finley (1979)
Liver	126.5	w	LD33	Red-winged blackbird	Finley (1979)
Liver	306 total/20.4 MeHg	d	No adverse effects observed	Black-footed albatross	Gochfeld (1980)
Brain	4-6	w	Failure to hatch	Black duck	Hoffman (1979)
Brain	20	w	25% mortality	Zebra finch	Scheuhammer (1988)
Egg	1-5/0.2-1.0	d	Reduced productivity in one half of the population	Merlin	Newton (1988)
Egg	0.5-1.5	w	Decreased hatchability	Pheasant	Heinz (1979)
Egg	0.86	w	Aberrant nesting behavior	Common loon	Heinz (1979)
Egg	1.0	w	Successful reproduction	Common tern	Finley (1978)
Egg	1.0-3.6	w	Residue threshold for significant toxic effects	Variety of water birds	Zillioux (1993)
Egg	2-16	w	No decreased hatchability	Herring gull	Finley (1978)
Egg	3.65	w	27% hatching, 10-12% fledging	Common tern	Finley (1978)
Kidney	37.4 total/6.2 MeHg	d	No adverse effect observed	Black-footed albatross	Kim (1996)
Kidney	40.4	w	LD33	Grackle	Finley (1979)
Kidney	74.3	w	LD33	Red-winged blackbird	Finley (1979)
Kidney	86.4	w	LD33	European starling	Finley (1979)

¹ LD33 = lethal dose, 33%

Relatively little information is available on the effects of Hg on amphibians. However, Bank et al. (2005) reported concentrations of total and MeHg in several northern two-lined salamanders (*Eurycea bislineata bislineata*) collected from streams in ACAD, the Bear Brook watershed, and SHEN. Total Hg levels in salamander larvae were significantly higher at sites in Maine, as compared with sites in SHEN. At the Bear Brook watershed, total Hg levels in salamanders were higher in the $(\text{NH}_4)_2\text{SO}_4$ treated subwatershed than in the reference subwatershed. Study results suggested that watershed features, including fire, $(\text{NH}_4)_2\text{SO}_4$ addition, wetland extent, and forest cover type affected Hg bioaccumulation in salamanders (Bank et al. 2005).

Four out of nine ponds in ACAD (44%) sampled by Bank et al. (2007b) had Hg methylation efficiencies of more than 10%, suggesting that palustrine food webs in ACAD are highly susceptible to bioaccumulation of MeHg. Total Hg in water was a strong indicator of both MeHg concentrations in the pond and total Hg in green frog (*Rana clamitans*) and bullfrog (*Rana catesbeiana*) tadpoles. One-third of sampled ponds had sediment Hg concentrations that approximated or exceeded the established effect thresholds for freshwater sediments. Mercury toxicity is of particular concern to amphibians, which cannot shunt toxics away from vital organs to body parts such as feathers, hair, or carapaces (as in the case of birds, mammals, and reptiles, respectively; Bank et al. 2006). Mercury concentrations in the two-lined salamander in ACAD were considered elevated, and tadpoles collected from streams in ACAD had Hg body burdens approximately threefold higher than green frog and bullfrog tadpoles. Because the salamander tadpoles are predators, whereas frog tadpoles are grazers, differences in Hg burdens may be due to differences in diet (Bank et al. 2007b). This puts two-lined salamanders and other higher-trophic level predators in ACAD at potential risk due to Hg toxicity. Bank et al. (2006) suggested that the effects of toxic Hg and AI may be the most likely causes of the dramatic and perhaps irreversible decline in ACAD of the northern dusky salamander, whose diet includes two-lined salamanders and other Hg-contaminated prey.

Davis (2013) investigated Hg methylation in vernal pools in NETN parks. These pools provide important breeding habitat for amphibians. Environmental conditions in the pools, including anoxic conditions at the sediment/water interface, low pH, and high DOC, contribute to enhanced methylation. He found high methylation efficiency (mean 43%; maximum 58%) at the eight study sites, suggesting high bioavailability of Hg to vernal pool biota.

Evers et al. (2005) compiled a database of over 4,700 records of avian Hg levels in the northeastern United States and eastern Canada. Using the belted kingfisher (*Ceryle alcyon*) and bald eagle (*Haliaeetus leucocephalus*) as indicators, they found increased Hg bioavailability from marine, to estuarine, to riverine systems. Bioavailability was highest in lakes. Differences in Hg exposure among species were correlated mainly with trophic position and availability of MeHg (Evers et al. 2005).

Studies of the transfer of Hg within food webs have mainly focused on freshwater aquatic ecosystems, which are considered to be at greatest risk of Hg biomagnification. Some studies have also been conducted on terrestrial upland ecosystems, including studies of Hg bioaccumulation in passerine birds. For example, Rimmer et al. (2005) documented MeHg availability in insectivorous passerines at 21 mountaintop locations in the northeastern United States. Mean blood Hg

concentration at breeding site locations varied from 0.08 to 0.38 µg/g (wet weight), and were highest in southern portions of the study area. Older male Bicknell's thrush (*Catharus bicknelli*) that breed in New England were judged to be at highest risk (Rimmer et al. 2005).

Rimmer et al. (2009) investigated food web transfer of Hg at high elevation in Vermont. Concentrations of Hg in Bicknell's thrush varied during the summer as the birds' diet shifted from detrital-based in early summer (higher Hg concentration) to foliage-based in late summer (lower Hg concentration). In general, Hg concentration increased in the following order: herbivore-detritivore-omnivore-carnivore. Raptors had higher blood Hg concentrations than their songbird prey (Rimmer et al. 2009).

There is a reasonable likelihood that high levels of MeHg, coupled with Ca depletion in acidified environments, is contributing to adverse impacts on terrestrial wildlife in the NETN. Evers et al. (2009) sampled invertebrates and breeding songbirds at 20 locations in New York and Pennsylvania in 2006. Forty seven bird species and 359 individuals were sampled to determine blood levels of Hg. Songbirds having highest blood Hg levels were generally bog-obligate species such as the palm warbler (*Dendroica palmarum*), upper canopy foragers such as the red-eyed vireo (*Vireo olivaceus*), and species commonly associated with riparian habitats (e.g., Louisiana waterthrush [*Seiurus motacilla*]). Blood Hg concentration varied across space and with elevation and likely also trophic level. Wood thrush (*Hylocichla mustelina*) was suggested as a good indicator of Hg impacts because they tended to have high blood Hg levels and because they have experienced substantial declines in recent years in distribution and abundance throughout New York and the Appalachian Mountains.

Hames et al. (2002) demonstrated a strong negative relationship between acidic deposition and wood thrush breeding, after accounting for several habitat variables. Breeding populations of wood thrush appeared to be most strongly affected in areas of low soil pH at high elevation where forest habitat was fragmented. It has been hypothesized that Hg is incorporated from leaf litter into invertebrates that feed on leaf tissue, and by insect predators, that are in turn preyed upon by song birds (Evers et al. 2009).

The snapping turtle (*Chelydra serpentina*) is particularly a good indicator of Hg contamination because of its omnivorous diet and long life span (Gibbs et al. 2007). It has previously been shown to accumulate toxics, including Hg, PCBs, and dioxins (Golet and Haines 2001, Bergeron et al. 2007). Turnquist et al. (2011) evaluated Hg concentration in snapping turtles sampled across New York. Fifth-eight turtles were studied at 10 lakes and wetlands. Total Hg concentration in muscle tissue was significantly correlated with lake SO_4^{2-} concentration and the maximum elevation of the watershed. The total Hg concentration of the shell was significantly correlated with water ANC, maximum elevation of the watershed, percent open water, lake-to-watershed area ratio, and various components of atmospheric Hg deposition (Turnquist et al. 2011).

Fish constitute an important pathway for transferring MeHg to wild mammals and birds. Monitoring of fish for Hg often focuses on both small prey fish and large, older piscivorous fish. The former are expected to respond quickly to changes in Hg exposure and reflect trophic transport of contaminants in food webs (Wiener et al. 2007). The latter respond more gradually to Hg bioavailability, and are

influenced by such factors as fish age, fish size, nutrient input, and inter-species competition (Mason et al. 2005). Thus, fish monitoring is often targeted at both the mid-trophic level omnivorous prey fish and the higher-trophic level predatory fish in order to capture the full range of fish Hg conditions (Kamman et al. 2005).

The concentration of Hg in fish tissue in the NETN is often positively correlated with lake and/or watershed area, and negatively correlated with pH, ANC, and zooplankton density (Chen et al. 2005, Driscoll et al. 2007b). Lake types that are generally associated with the most Hg bioaccumulation are poorly buffered, low in pH and productivity, and have forested watersheds and little human development within the watershed (Chen et al. 2005). The review of Evers (2005) on Hg pollution in the Northeast classifies Hg-sensitive surface waters as those having high SO_4^{2-} concentrations, low pH and ANC, extensive wetlands, large watershed area relative to lake area, fluctuating water level, and low nutrient concentration.

Several piscivorous bird and mammal species have been suggested as biomonitors of Hg bioaccumulation in the NETN (Wolfe et al. 2007). In particular, the common loon and bald eagle are good indicators of Hg risk to wildlife (Evers 2006). Increased concentrations of Hg have been found to be associated with behavioral, physiological, and reproductive effects on these bird species (Burgess and Meyer 2008, Evers et al. 2008).

A study by Longcore et al. (2007b) compared Hg contamination in tree swallows in ACAD with tree swallows at an Hg-contaminated EPA Superfund site in Ayer, MA. They determined that MeHg concentrations in feathers of birds living at Aunt Betty Pond in ACAD were higher than in birds at the Superfund site. Eggs from both locations exceeded the embryotoxicity threshold for Hg. Swallows with high Hg burdens in their feathers may expose raptors and other high trophic-level predators to concentrated and potentially toxic levels of Hg. For example, peregrine falcons (*Falco peregrines*) nesting in ACAD may feed on Hg-contaminated songbirds (Longcore et al. 2007a). Additionally, the authors suggested that Hg residues in tree swallow tissues in the northeastern United States were higher than those of birds from the Midwest, perhaps due to greater local sources of Hg, higher Hg atmospheric deposition rates, and more prevalent hydrological conditions that promote the conversion of inorganic Hg to MeHg (Longcore et al. 2007b).

Although the fauna in ACAD show elevated tissue Hg concentrations, the physiological and ecological implications of this Hg contamination are unclear (Vaux et al. 2008). Exposure of park fauna to Hg pollution likely represents a moderate to high risk (Bank et al. 2007a). Continued study of potential adverse impacts are needed. Identified priority research needs at ACAD include continuation of long-term air monitoring, research to establish CLs, studies to identify biological and ecological endpoints associated with pollutant exposure, and process-based Hg experiments (Maniero and Breen 2004, Tonnessen and Manski 2007).

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