

Lakewater chemistry at Acadia National Park, Maine, in response to declining acidic deposition

J.S. Kahl, Senator George J. Mitchell Center for Environmental and Watershed Research, 5710 Norman Smith Hall, University of Maine, Orono, ME 04469, kahl@maine.edu

S.J. Nelson, Senator George J. Mitchell Center for Environmental and Watershed Research, 5710 Norman Smith Hall, University of Maine, Orono, ME 04469, sarah.nelson@umit.maine.edu

J.L. Stoddard, Western Ecology Division, U.S. Environmental Protection Agency, 200 S.W. 35 Street, Corvallis, OR, stoddard.john@epa.gov

S.A. Norton, 111 Bryand Global Sciences Center, University of Maine, Orono, ME 04469, norton@maine.edu

T.A. Haines, 313 Murray Hall, University of Maine, Orono, ME 04469, haines@maine.edu

Abstract. Surface water chemistry in the northeastern U.S. has changed significantly as a result of changes in atmospheric deposition during the past 20 years. During 1990-2000, sulfate concentrations in deposition declined significantly at a rate between -0.75 and -1.5 $\mu\text{eq/L/year}$. Nitrogen concentrations (nitrate + ammonium) declined slightly. Base cations in deposition have not changed significantly. These changes in deposition are a continuation of trends that have been occurring for more than 20 years. In response to the changes in deposition, sulfate concentrations in sensitive Maine lakes have declined by 10 to 25% since 1982, in part due to the 1995 implementation of Phase I controls of the Clean Air Act Amendments and in part to other long-term pollution reduction efforts. Sulfate in surface waters of Acadia National Park declined 10%, the low end of the range in the region, in part because of the input of sulfate from marine aerosols. The expectation for reductions in sulfate was a parallel reduction in lake acidity. However, the slight trend toward decreased lake acidity seen in the 1980s in Maine has slowed or reversed in the 1990s. As a group, lakes at Acadia show a slight decrease in acidity. The reason for the lack of recovery in the region is the simultaneous decline in base cation concentrations that are a by-product of alkalinity generation, e.g. calcium (Ca), magnesium (Mg). At Acadia, the decline in the sum of Ca + Mg was 14%. The decline in base cations appears to be related to several interacting factors: continued high atmospheric deposition of nitrogen, a lag time in response, or the influence of climate and acidic deposition on watershed response. Freshwaters at Acadia may hold a key to understanding the regional response in base cations, because deposition of base cations from the atmosphere is very high in this island environment (especially magnesium), compared to other regions with long-term water chemistry data.

1. Introduction

Title IV of the 1990 Clean Air Act Amendments (CAAA) in the U.S. set target reductions for sulfur and nitrogen emissions from industrial sources to reduce the acidity in deposition. These reductions have continued the trend of reductions in emissions and deposition of sulfur during the past 30 years, with the rate of decline accelerated by Phase

I of the 1990 CAAA that was implemented in 1995 (Lynch *et al.*, 2000). Slight reductions in nitrogen emissions have occurred since 1996.

One of the intended effects of the CAAA reductions was to decrease the acidity of low alkalinity waters and thereby improve their biological condition. The key science and policy questions related to the CAAA are 1) whether the declines in emissions yield reductions in acidic deposition; 2) whether changes in deposition cause changes in surface water chemistry; and, 3) whether biologically-relevant water chemistry has improved in acid sensitive regions as a result of changes in deposition.

Documentation of acidification of surface waters began in Scandinavia (e.g., Oden, 1968) although reports of acidic lakes date back to the 1950s in North America (Gorham, 1957). Recognition of the issue became common in the U.S. in the early 1970s (Likens *et al.*, 1972), with identification of impacts on fish by the mid-1970s (e.g., Schofield, 1976). Trend assessments for surface waters have been common in the literature for more than a decade, with the general conclusion that surface water recovery is slow to non-existent (Stoddard and Kellogg, 1993; Webster *et al.*, 1993; Kahl *et al.*, 1993; Driscoll and van Dreason, 1993; Dewalle and Swistock, 1994; Driscoll *et al.*, 1995; Likens *et al.*, 1996; Mattson *et al.*, 1997; Stoddard *et al.*, 1998; Driscoll *et al.*, 2001; Skjelkavale *et al.*, 2001; Evans and Monteith, 2001; Stoddard *et al.*, 2003).

This paper reports on lake chemistry data collected at Acadia National Park since 1982 (Kahl *et al.*, 1985; Figure 1), and puts the response to acidic deposition in the context of the recent assessment of the response of surface waters in the northeastern US to changes in atmospheric deposition (Stoddard *et al.*, 2003).

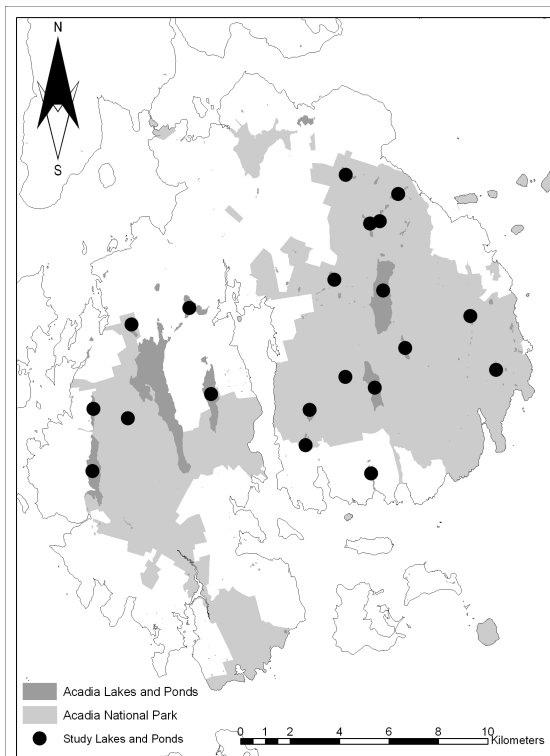


Figure 1. Location of Acadia National Park on Mount Desert Island in Maine, USA with locations of study lakes and ponds.

Why do we care about lake and stream chemistry? Long-term chronic acidification and short-term temporary episodic acidifications are of concern in regions receiving acidic deposition. Surface water chemistry is a direct indicator of the potential deleterious effects of acidification on biotic integrity. Because surface water chemistry integrates the sum of processes upstream in a watershed, it is also an indicator of the *indirect* effects of watershed-scale impacts, such as nitrogen saturation, forest decline, or soil acidification.

Biologically-relevant surface water chemistry. The main cause for concern over the effects of surface water acidification in the U.S. and elsewhere is the potential for detrimental biological affects (Baker and Christensen, 1991; National Atmospheric Deposition Program (NADP), 1998). Typically, there is concern for biological impact if surface water pH is less than 6. At low pH values, aluminum may be present at concentrations that are toxic to biota, including sensitive life stages of fish and sensitive invertebrates. Aluminum is an abundant and normally harmless component of rocks and soils. However, it leaches from silicate minerals when they come in contact with low pH waters. While much of the aluminum present in surface waters is organically-bound and relatively non-toxic, certain inorganic species are highly toxic.

Nitrogen saturation. One of the key remaining research issues is the role of nitrogen in watershed responses to acidic deposition. The concept of *nitrogen saturation* (Aber *et al.*, 1989; Stoddard, 1994) received increasing attention in the 1990s (Mitchell *et al.*, 1996; Williams *et al.*, 1996; Aber *et al.*, 1998). Nitrogen (N) saturation is defined as deposition of N to a watershed in excess of the assimilative capacity of soils and vegetation, resulting in the export of nitrate (NO₃). Nitrate export can contribute to acidification (especially episodic acidification), mobilization of aluminum, and leaching of cations from soils (Aber *et al.*, 1998).

2. Methods

This paper uses long-term records of wet deposition from the National Atmospheric Deposition Program (NADP (<http://nadp.sws.uiuc.edu/>), repeat surveys of lake chemistry at Acadia National Park (Acadia NP) dating back to 1982 (Kahl *et al.*, 1985; Kahl, 1996), data collected by the Resource Management Division of Acadia NP, and long-term records of lake chemistry from research conducted in New England (Kahl *et al.*, 1991; Stoddard *et al.*, 2003). The lake data from Acadia NP cover the period from 1982 to either 1998 or 2000, depending on the availability of data at each site. Accepted EPA methods and quality assurance are documented in various publications (Nelson and Kahl, 2003; Morrison, 1989, Newall *et al.*, 1987; Hillman *et al.*, 1986).

Results and Discussion

Declines in sulfate deposition. Sulfate concentrations declined substantially in the Northeastern US at a median rate between -1.0 and -1.5 $\mu\text{eq/L/year}$ for the period 1990 to 2000 (Table 2). At the Acadia NP NADP site, the rate of change in sulfate was -0.53 $\mu\text{eq/L}$. Changes in sulfate emissions correspond directly to changes in sulfate deposition.

Declines in nitrate emissions and deposition. Decreases in NO_x emissions were more modest than those of sulfur (Table 2). There was a slight increase in N deposition at Acadia NP. Since 1990, total utility NO_x emissions (Phase I and II sources) were reduced an average of 23% nationally, following implementation of Phase I of the Acid Rain Program. However, electric utilities contribute only about one-third of total NO_x emissions. Total NO_x emissions from other sources have remained relatively constant (motor vehicles and other industrial sources also contribute significantly), and therefore the reductions achieved under the Acid Rain Program have not resulted in a significant change in total NO_x emissions and deposition.

Increases in pH and base cations in deposition. Lynch *et al.* (2000) found significant declines in hydrogen ion at many NADP stations during 1990-2000, at rates that were less than the decrease in sulfate. Wet deposition of hydrogen ion decreased in every region (Table 2). Base cation deposition increased non-significantly in the northeastern US, continuing the pattern of flat to increasing base cation deposition at most stations in the region during the past 20 years. Deposition of base cations increased significantly at Acadia NP, driven by decadal increases in deposition of marine salts from the Gulf of Maine (Figure 1).

Status of surface waters. The National Surface Water Survey (NSWS) documented the status and extent of chronic acidification during probability surveys conducted from 1984 through 1988 in acid sensitive regions throughout the U.S. (Linthurst *et al.*, 1986; Landers *et al.*, 1988; Kaufman *et al.*, 1988). The NSWS concluded that 4.2% of lakes in the northeastern US were acidic. The Adirondack Mountain region had the greatest proportion of acidic surface waters (14%) for lakes larger than 4 hectares. Counting smaller lakes, the Adirondack Lake Survey Corporation estimated that 26% of lakes larger than 0.5 hectares were acidic (Driscoll *et al.*, 1991). The large numbers of lakes in these regions translate to several hundred acidic waters in each region. At Acadia NP, Sargent Mountain Pond and Duck Pond are acidic, representing 9% of the 21 lakes sampled. This is a higher percentage than in the rest of Maine, reflecting the granitic bedrock and thin soils common at Acadia NP.

Table 1. Regional trend results (1990-2000) for atmospheric deposition (wet-only annual concentration data from NADP/NTN network) in acid sensitive regions (from Stoddard *et al.*, 2003). All units are $\mu\text{eq/L/yr}$. Values are the median slopes for each region, with significance determined by calculating confidence intervals around each regional median. Data for Acadia NP are the NADP results for the station at McFarland Hill (NADP, 2002). Base cations are defined here as the sum of Ca + Mg.

Region	SO ₄	Nitrogen	NO ₃	Base Cations	Hydrogen Ion
New England	-0.96**	-0.26*	-0.20**	+0.02 ^{ns}	-0.81**
Adirondacks	-1.47**	-0.37**	-0.38**	+0.01 ^{ns}	-1.48**
Acadia NP	-0.53**	+0.04 ^{ns}	+0.05 ^{ns}	+0.17**	-0.29*

^{ns} regional trend not significant ($p > 0.05$)

* $p < 0.05$

** $p < 0.01$

Changes in surface water chemistry. Our analysis of surface water response to changing deposition focuses on the key variables that play major roles in acidification and recovery: sulfate and nitrate, base cations, pH and ANC (acid neutralizing capacity), and DOC (dissolved organic carbon), a possible indicator of changes in natural organic acidity.

Sulfate (SO₄) declined in surface waters in the glaciated regions of the northeastern US by median values between -2 and -4 µeq/L/year (-0.4 µeq/L at Acadia NP; Table 3). The declines in SO₄ concentrations are almost certainly direct responses to declining emissions and SO₄ deposition in the 1990s, and represent the most dramatic effects of Title IV of the CAAA and previous emissions regulations. These changes in emissions and deposition continue the trend in declining SO₄ that has been occurring for three decades (Stoddard *et al.*, 2003).

Stoddard *et al.* (2003) concluded that surface waters in glaciated terrain have, on average, responded relatively rapidly to the decline in sulfate deposition. Additional reductions in deposition will result in additional declines in surface water concentrations of sulfate in glaciated terrain.

Changes in NO₃ were much smaller than changes in SO₄, with the only significant changes occurring in the two regions with the highest ambient NO₃ concentrations. Most waters at Acadia NP have low NO₃ concentrations. However, sites such as the PRIMENet sample site at Hadlock Brook have significant leakage of NO₃, and there is no indication in the data that NO₃ concentrations have declined at Hadlock Brook as they have in many other areas during the 1990s. These unexplained changes in NO₃ in many surface water concentrations at a time of stable N deposition underscore the complexities of N biogeochemistry. We expect that that a decline in N deposition will lead to general declines in surface waters, but the timing and correlation cannot be predicted at this time.

Table 2. Regional trend results for Long-Term Monitoring sites for the period 1990 through 2000 (Stoddard *et al.*, 2003). Values are median slopes for set of sites in each region. Units for sulfate, nitrate, base cations [Ca + Mg], Gran ANC and hydrogen are µeq/L/year. Units for DOC are mg/L/year (insufficient historical data at Acadia NP). Units for aluminum are µg/L/year.

Region	SO ₄	NO ₃	Base Cations	Gran ANC	Hydrogen	DOC	Aluminum
New England Lakes	-1.77**	+0.01 ^{ns}	-1.48**	+0.11 ^{ns}	-0.01 ^{ns}	+0.03*	+0.09 ^{ns}
Adirondack Lakes	-2.26**	-0.47**	-2.29**	+1.03**	-0.19**	+0.06**	-1.12**
Acadia lakes	-0.39*	-0.06 ^{ns}	-0.43*	+0.33 ^{ns}	+0.02 ^{ns}	NA	+0.05 ^{ns}

^s regional trend not significant (p > 0.05)

* p < 0.05

** p < 0.01

NA insufficient data

Increasing ANC is the main indicator of recovery from acidification. In the northeastern US, there were modest increases in the Adirondack region and at Acadia NP (Table 3). Hydrogen ion (acidity) followed ANC with small declines in each region.

The largest recovery should be possible at sites that have undergone the most severe acidification. Stoddard *et al.* (2003) analyzed Gran ANC trends by ANC class and determined that the most impacted sites recovered faster in the 1990s. Sites with ANC less than zero gained ANC four times faster than sites with ANC greater than 25 $\mu\text{eq/L}$. The average increase in ANC was 12% for acidic lakes, 7% for low ANC lakes, and less than 1% for lakes with ANC greater than 25 (Figure 2), suggesting that ANC values are converging during the recovery process toward an ANC that may be in the range of 25 to 30 $\mu\text{eq/L}$ ANC (Stoddard *et al.*, 2003).

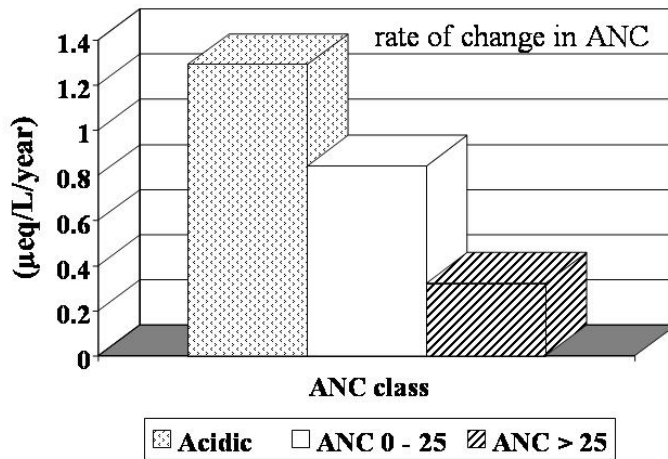


Figure 2. The lowest ANC (highest acidity) lakes in the northeast are responding fastest to declining acidic deposition.

One of the most universal watershed responses to acidic deposition is the mobilization of base cations from soils. As rates of acidic deposition decline, and the supply of acid anions to watersheds soils decreases, the rates of cation mobilization are also expected to decrease. Lowered rates of cation mobilization translate to declines in surface water base cation concentrations, a change widely observed in the northern hemisphere for more than a decade. All of the glaciated regions in the northern and eastern U.S. exhibited significant declines in base cation [Ca + Mg] concentrations in the range of -1.5 to -2.5 $\mu\text{eq/L/year}$. This decline in base cations offsets some of the decline in sulfate concentrations, and limits the extent of recovery.

4. Conclusions

The rate of change in surface water ANC appears to largely be the result of changes in acid anions versus base cations, as represented by:

$$\text{Change in ANC} = \text{change in } [\text{Ca} + \text{Mg} + \text{Na} + \text{K}] \text{ minus change in } [\text{SO}_4 + \text{NO}_3 + \text{Cl}]$$

Regionally, SO_4 has decreased at a rate of approximately -2.5 $\mu\text{eq/L/year}$ (the mean of regional median slopes), and NO_3 at a rate of -0.5 $\mu\text{eq/L/year}$, in surface waters of glaciated terrain. These rates of change set an upper limit to our expectation of ANC

recovery of +3 $\mu\text{eq/L/yr}$ (i.e., the sum of declines in SO_4 and NO_3). The actual increase in Gran ANC is about +1 $\mu\text{eq/L/year}$, because the decline in SO_4 and NO_3 (acid anions) has been offset by a decline of 1.8 $\mu\text{eq/L/year}$ in base cations. At some sites, the decline in base cations has exceeded the decline in acid anions, and these sites have acidified.

This general pattern occurs in the lakes at Acadia NP. For example, Ca + Mg in Bubble Pond declined at a rate of 0.6/year $\mu\text{eq/L}$ while sulfate was declining faster, at 1.2 $\mu\text{eq/L/year}$. The difference is a 0.6 $\mu\text{eq/L/year}$ increase in ANC, exactly the rate of increase in the data. Conversely, The Bowl has acidified slightly because base cations decreased faster (-0.6 $\mu\text{eq/L/year}$) than the acid anions (-0.3 $\mu\text{eq/L/year}$).

Regionally, there has been some recovery in ANC, especially in the lowest ANC waters. Stoddard *et al.* (2003) estimated that there are about one third fewer acidic lakes (ANC < 0) in the Northeast during the past 15 years, although these lakes still have very low ANC. At Acadia NP, Sargent Mountain Pond and Duck Pond both have higher ANC (less acidity) in 2000 compared to 20 years ago, but both are still acidic (i.e., negative ANC).

We do not know if the rates of increase in ANC will continue without further reductions in deposition. This is the major uncertainty for rates of recovery: to what extent is recovery resulting from recent changes in deposition already reflected in current surface water chemistry, or are further reductions in deposition necessary to continue the present trends? The only way to answer this question is to maintain the commitment to long-term assessments of surface water chemistry in regions such as Acadia NP that have waters that are sensitive to the effects of acidic deposition.

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