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Response of Watersheds in the Catskills and Adirondacks to Reduced Atmospheric Acid Inputs

Report Summary

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Report Summary

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Abstract

Regional and national reductions in emissions of sulfur and nitrogen oxides have resulted in large decreases in acid deposition to forest ecosystems in the Catskills and Adirondacks of New York State. Nevertheless, soils in these areas continue to acidify and the recovery of acid neutralizing capacity (ANC) in surface waters has been slow. Data from 26 headwater streams in the Catskills and inlet streams at three Adirondack lakes all show large decreases in sulfate concentrations between 1994-2000 and 2010-2013, a direct consequence of lower acid inputs. These declines were largely offset by decreases in stream calcium (Ca) concentrations, resulting in little or no change in ANC. In high-ANC Catskills streams, decreases in Ca concentrations exceeded decreases in sulfate, suggesting decreases in ANC (Section 2). In the Adirondack lake inlets, changes in ANC were generally small (Section 3). In both regions, decreases in solute concentrations have resulted in lower ionic strength in surface waters, which could be significant ecologically and, in the case of the Catskills, for public water supply.

Decreases in surface-water Ca concentrations have been ascribed to the depletion of Ca from soils in regions impacted by chronic acid deposition. Data from 25 headwater catchments in the Catskills indicate that soils have very low concentrations and pools of exchangeable Ca, Mg and K (Section 4). In particular, pools of soil Ca are especially low relative to stream Ca fluxes and Ca pools in forest vegetation, calling into question the long-term sustainability of upland Catskills forests. Similarly, results from computer modeling of biogeochemical processes in Adirondack lake watersheds indicate that the ANC in some lakes is unlikely to rise to the level necessary to support a viable sport fishery even with a 100% reduction in sulfur deposition (Section 5). However, those simulations also suggest that adoption of a total maximum daily load (TMDL) of $1.47 \text{ kg S ha}^{-1} \text{ yr}^{-1}$ could result in substantial improvement in Adirondack lakes that currently have ANC concentrations below $20 \mu\text{eq L}^{-1}$.

Keywords

Acid rain, acidification, Adirondacks, calcium, Catskills, critical loads, modeling, soil acidity, soil chemistry, stream water, total maximum daily loads

Acknowledgments

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Acronyms and Abbreviations

μeq	microequivalents
μmol	micromole
ALSC	Adirondack Lake Survey Corporation
ANC	acid neutralizing capacity
Ca	calcium
K	potassium
L	liter
Mg	magnesium
N	nitrogen
Na	sodium
S	sulfur
SO_4^{2-}	sulfate
TMDL	total maximum daily load
yr	year

1 Background

The 1970 Clean Air Act and subsequent air quality rules have resulted in substantial reductions in acidic deposition, both sulfuric and nitric acid. This environmental success story is tempered somewhat by the relatively slow recovery of the acidity of surface waters in regions historically impacted by acid deposition. In the Adirondack region of New York, for example, concentrations of the acid anions sulfate and nitrate (NO_3^-) in precipitation have declined markedly between 1978 and the present, yet the acid neutralizing capacity (ANC) in Adirondack lake waters has increased by only $0.95 \pm 0.32 \mu\text{eq L}^{-1} \text{yr}^{-1}$ over a similar time period (Driscoll et al., in review). This slow recovery of surface-water acidity has been attributed to decreases in basic cation concentrations, particularly calcium (Ca^{2+}), possibly related to the depletion of Ca from forest soils (Driscoll et al. 2001, Warby et al. 2005, Driscoll et al. in review). Indeed, long-term soil studies have documented large, statistically significant decreases in soil exchangeable Ca in the Adirondack region (Johnson et al. 2008, Sullivan et al. 2006, Warby et al. 2009) and elsewhere (Bailey et al. 2005, Warby et al. 2009).

Constable Pond in the Adirondacks



Watersheds in the Adirondack and Catskills regions of New York have historically received atmospheric acid inputs among the highest in the United States (Ollinger et al. 1993). Surface waters in both regions have relatively low Ca concentrations, and there is some evidence that Ca levels have decreased (Lawrence et al. 1999, Warby et al. 2009). Although some soil data are available for the Adirondacks, the absence of sufficient baseline soil chemical data in the Catskills has made it impossible to evaluate the degree to which changing Ca concentrations in streams may be related to exchangeable Ca in soils. The history of acid deposition in the region, coupled with its role as the principal water supply to New York City, makes the assessment of changes in soil and surface-water chemistry essential to future science-based policymaking.

Work related to the recovery of aquatic systems in the Adirondacks has focused on lake watersheds due to their abundance in the region and their cultural, recreational, and economic importance. With a few notable exceptions, stream systems have received less attention in the Adirondacks. Monitoring of additional stream sites in the region can provide crucial data for (1) the assessment of the recovery of running waters from decades of chronic acidification; and (2) better understanding the inputs to sensitive lakes in the region.

In this project, the chemistry of 26 headwater streams in the Catskills and the inlets to three lakes (Grass Pond, G Lake, and Constable Pond) in the Adirondacks were measured to assess their recovery under reduced acid deposition. This work builds upon data collected in the 1990s by the Cary Institute of Ecosystem Studies (Lovett et al. 2000) and by the Adirondack Lake Survey Corporation (NYSERDA 2011; Ito et al. 2007). We also collected and analyzed soils from 25 Catskills watersheds to establish a soil chemical baseline for headwater catchments in the region. Finally, these and other data were used in a comprehensive biogeochemical model to estimate total maximum daily loads (TMDLs) for sensitive lake-watersheds in the Adirondack region.

2 Deionization of Catskills Streams

Stream waters were sampled from 26 headwater catchments across the Catskills that had previously been studied by Lovett et al. (2000). The samples were collected monthly from June 2010 to November 2012 and bimonthly from January to July 2013. In all 26 streams, Ca^{2+} was the dominant cation and sulfate (SO_4^{2-}) was the dominant anion, both in the 1990s and in this study. Results for an acidic (low-ANC) and a non-acidic (high-ANC) stream are shown in Figures 1 and 2, respectively. In both cases (and at the other sites as well), sulfate concentrations declined substantially between the 1990s and 2010-2013, corresponding to decreases in sulfate deposition and, ultimately, national sulfur emissions. Nitrate concentrations, which are highly seasonal due to biological nitrogen (N) cycling, did not exhibit significant decreases despite declines in N deposition.

Figure 1. Water chemical trends in Prediger Brook, a low-ANC stream in the Catskills region

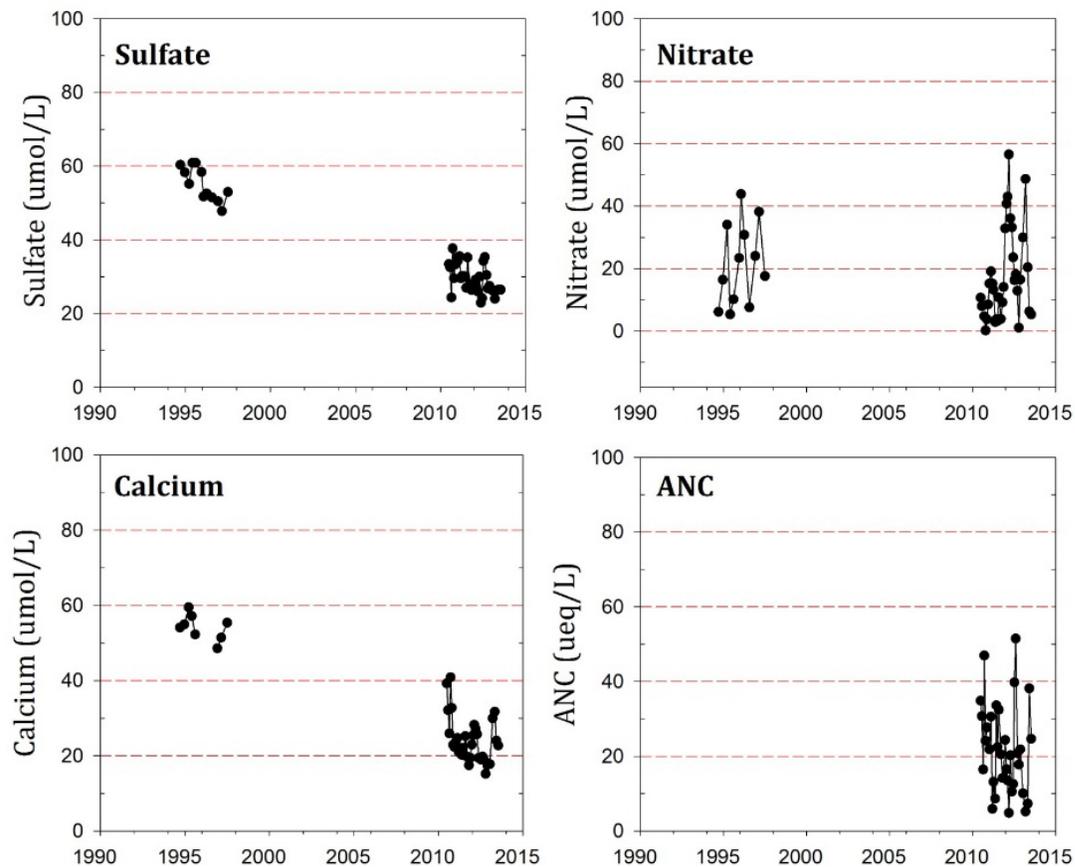
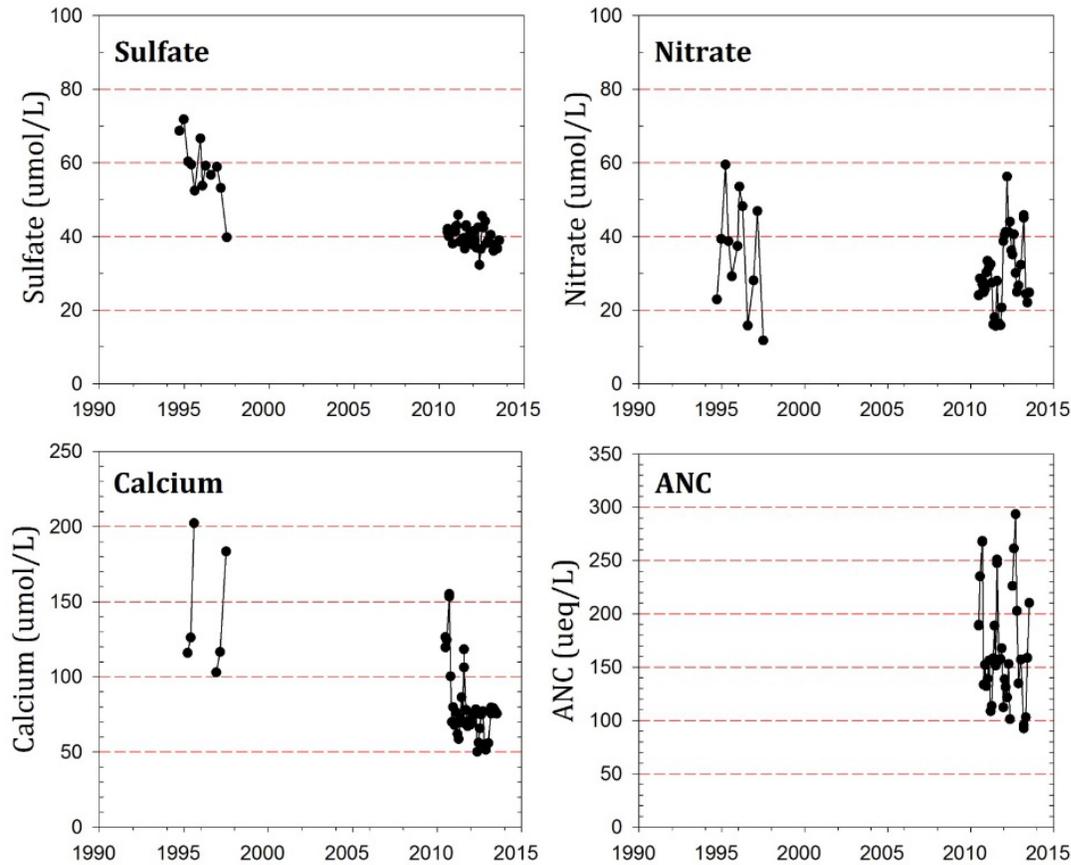


Figure 2. Water chemical trends in Mill Brook, a high-ANC stream in the Catskills region



Calcium concentrations in Catskills waters also declined sharply between the 1990s and 2010-2013. In low-ANC streams, the decline in Ca was proportionally large and approximately equal on a molar basis to the decline in sulfate. At Prediger Brook (Figure 1), for example, the decline in Ca concentration was approximately 30 $\mu\text{mol/L}$, a drop of more than 50% in 15 years and quantitatively equivalent to the decrease in sulfate concentration. In high-ANC streams, declines in Ca concentrations were greater than in low-ANC streams and in many cases exceeded the declines in sulfate. At Mill Brook (Figure 2), average Ca concentrations decreased by approximately 60 $\mu\text{mol/L}$, while sulfate concentrations decreased by only 20 $\mu\text{mol/L}$.

Buttermilk Falls in the Catskills



Acid neutralizing capacity was not specifically measured at these sites in the 1990s. However, ANC can be expressed as the difference between the concentrations of strong base cations (Ca^{2+} , Mg^{2+} , Na^+ , K^+) and strong acid anions (SO_4^{2-} , NO_3^- , Cl^-) as shown in Equation 1:

Equation 1 $\text{ANC } (\mu\text{eq/L}) = (2[\text{Ca}^{2+}] + 2[\text{Mg}^{2+}] + [\text{K}^+] + [\text{Na}^+]) - (2[\text{SO}_4^{2-}] + [\text{NO}_3^-] + [\text{Cl}^-])$

where the concentrations on the right side are expressed in $\mu\text{mol/L}$. Because Ca^{2+} and SO_4^{2-} are the dominant cation and anion, respectively, in these waters, the ANC change is likely driven by the changes in their concentrations according to Equation 2:

Equation 2 $\Delta\text{ANC} \approx 2(\Delta[\text{Ca}^{2+}] - \Delta[\text{SO}_4^{2-}])$

In low-ANC streams, the approximately equal decreases in Ca and sulfate concentrations suggest that changes in ANC were small. In high-ANC streams, where decreases in Ca concentrations exceeded decreases in sulfate, it is likely that ANC decreased.

The large decreases in the two main ions in Catskills waters indicate that the ionic strength of waters in the region is decreasing. Waters with low ionic strength are highly corrosive, a threat to water distribution systems and the intestinal tracts of animals. The deionization of Catskills waters, if it continues, may therefore be a significant concern for the New York City drinking water supply and to wildlife management in the region.

3 Acid Neutralizing Capacity (ANC) in Inlet Streams of Adirondack Ponds

The inlet streams in three Adirondack lake watersheds – Constable Pond (3 major inlets), Grass Pond (7 major inlets), and G Lake (4 major inlets) – were sampled from 2011 to 2013. The ALSC had previously sampled these sites from 1998 to 2001 (Ito et al. 2007). Results from this project showed some interesting patterns in stream chemistry, demonstrating the value of using inlet streams for monitoring the recovery of Adirondack watersheds from chronic acidification. As in the Catskills streams, substantial declines in sulfate and calcium concentrations were observed between 1998-2000 and 2011-2013 (Figure 3). However, unlike the Catskills sites, there is no apparent relationship between the decreases in calcium concentrations observed in these inlet streams and the decreases in sulfate (Figure 4), suggesting that declines in base cation deposition in precipitation cannot fully explain the drop in stream calcium.

Grass Pond in the Adirondacks



Little change was observed in ANC in two inlet streams at Grass Pond that have relatively high ANC (G3 and G4; Figure 3). However, in several streams characterized by near-zero or negative ANC, declines in ANC were observed from 1998-2000 to 2011-2013 (Figure 3). All four of the major inlet streams to G Lake exhibited lower ANC in 2011-2013 than 12 years prior, with decreases ranging from 8 to 42 $\mu\text{eq/L}$ (Figure 3). The decreasing ANC that was observed in low-ANC streams is inconsistent with the regional

trend in lake waters. For example, the average ANC in waters leaving G Lake increased from 1.7 to 13.9 $\mu\text{eq/L}$ between 1993 and 2009 (NYSERDA 2011), though the temporal pattern is subtle (Figure 5). This apparent disconnect between stream and lake trends indicates the potential importance of in-lake ANC generation to ANC export from Adirondack lake ecosystems.

Figure 3. Changes in sulfate, calcium and acid neutralizing capacity (ANC) in inlet streams to three Adirondack lakes

Bar heights indicate mean concentrations for 1998-2000 and 2010-2012. Error bars show one standard deviation.

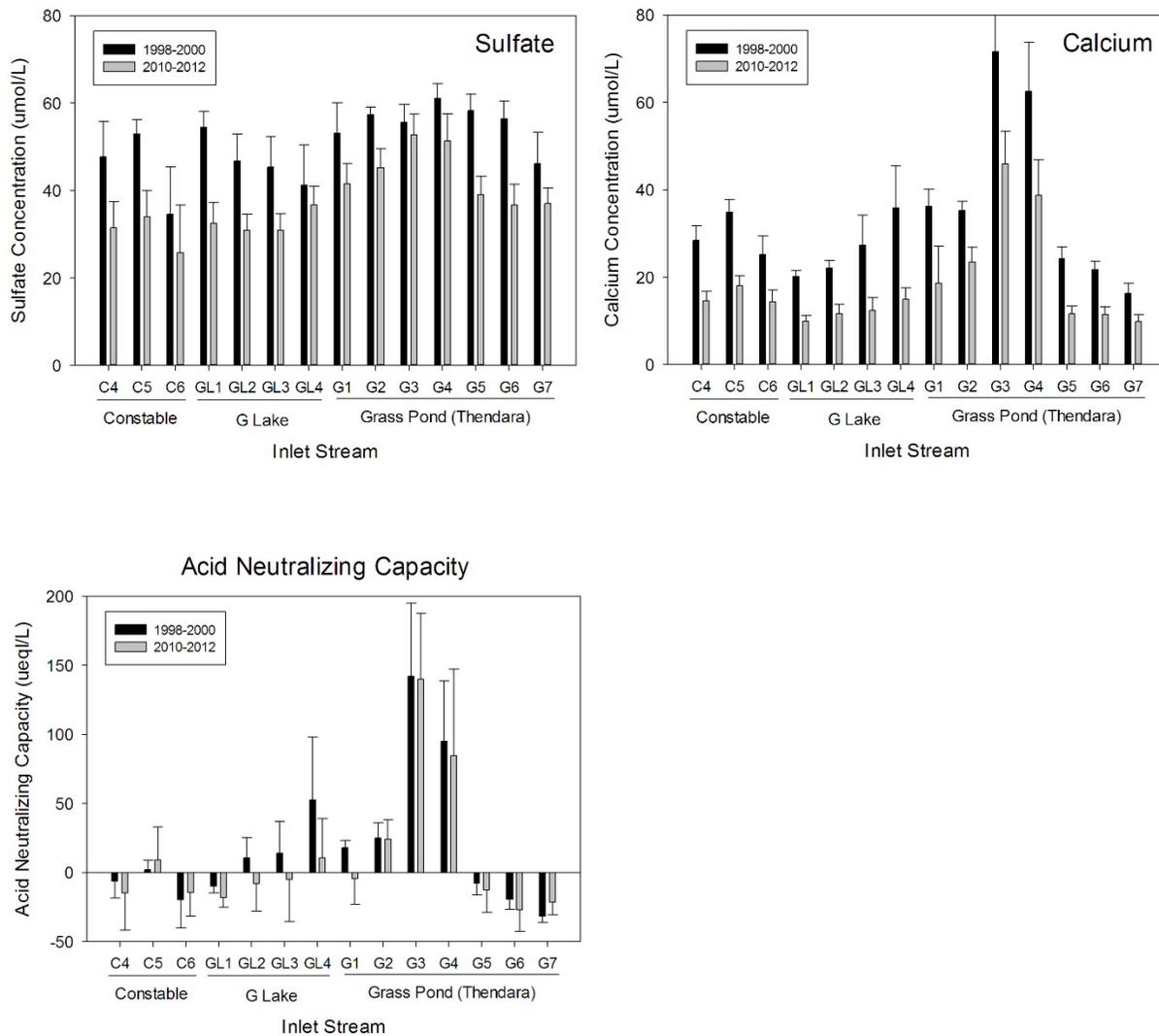


Figure 4. Decreases in calcium concentrations in inlet streams to three Adirondack lakes between 1998-2000 and 2011-2013 are unrelated to the decrease in sulfate concentration

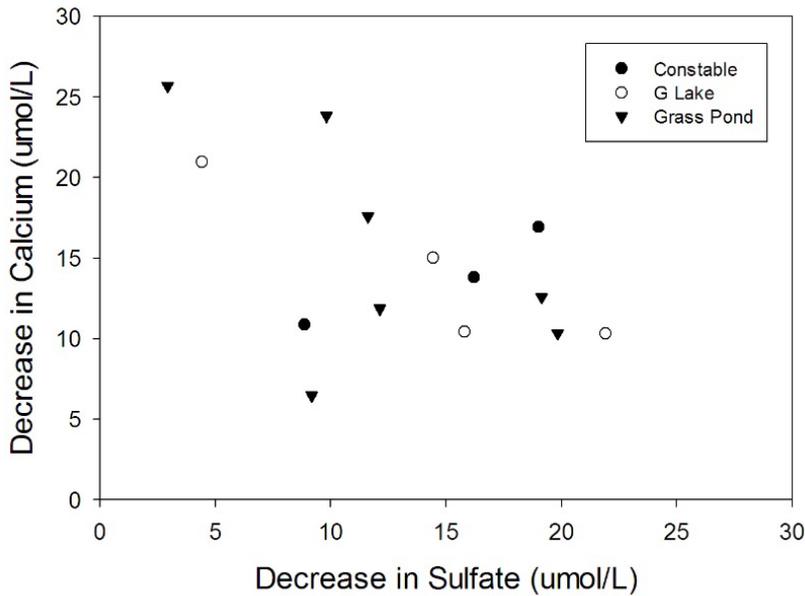
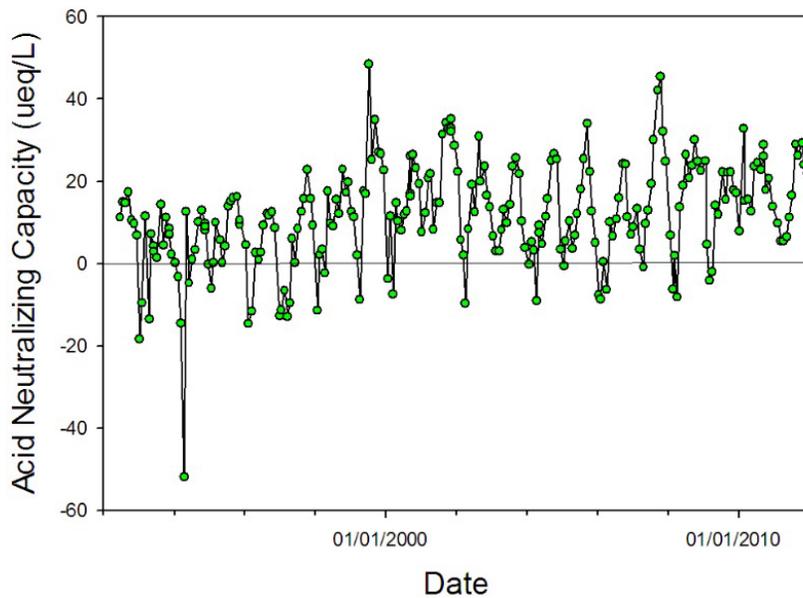


Figure 5. Long-term trends in acid neutralizing capacity (ANC) in waters draining G Lake in the Adirondacks

Average ANC increased from 1.7 $\mu\text{eq/L}$ to 13.9 $\mu\text{eq/L}$ between 1993 and 2009.

Data from Adirondack Lakes Survey Corporation



These inlet streams drain headwater watersheds with varying size, topography, and land cover attributes. The 14 inlets sampled at Grass Pond, G Lake, and Constable Pond range in area from 10-450 hectares, with wetlands representing up to 17% of total watershed area (Ito et al. 2007). Thus, the data capture a diverse population of stream watersheds representative of the Adirondack region. By monitoring lake inlet streams, the data generated can be used to inform modeling and policymaking related to lake ecosystems, which are culturally and economically significant in the Adirondack region. Although data indicate some improvements in water quality, the failure of ANC to increase in Adirondack watersheds continues to be a puzzle and a challenge for science-based ecosystem management in the region.

4 Chemistry of Upland Soils in the Catskills

In the Catskills, forest soils provide important services including maintaining excellent water quality. Soil processes in the region's forests maintain the purity of the New York City water supply, which currently does not require filtration prior to human consumption. This high quality persisted despite high inputs of acid deposition in the second half of the 20th century. The long-term declines observed in stream Ca concentrations may be the result of depleted soil exchangeable Ca, which has been observed in several studies in forest soils of the northeastern United States (Bailey et al. 2005; Johnson et al. 1994, 2008; Warby et al. 2009). These studies also found significant increases in exchangeable acidity, Al in particular. Therefore, the observation of decreasing soil Ca between the mid-1980s and 2000s indicates that forest soils of the region have continued to acidify during a period of sharp decreases in acid inputs, thus delaying the recovery of associated surface waters. Unfortunately, there have been no geographically extensive studies of soil chemistry in Catskills forests that could be used to assess soil change due to acid rain and recovery. Soils in 25 headwater catchments (coinciding with the stream sampling sites discussed in section 2) were sampled to create a baseline data set on forest soil chemistry in the Catskills (Figure 6).

A "quantitative pit" method was used (Figure 7) to sample soils, which provides a direct measurement of the soil mass per unit area for five layers: O_i+O_e horizon, O_a/A horizon, 0-10 cm mineral soil, 10-20 cm mineral soil, and 20 cm to C horizon or bedrock. Soil nutrient pools were calculated by multiplying measurements of soil chemical concentrations and soil layer masses (Table 1).

Figure 6. Soil samples were collected from 50 soil pits, two in each of 25 forest watersheds in the Catskills

This map shows the locations of the pits superimposed on a geologic map of the area.

Reprinted with permission from Johnson 2013.

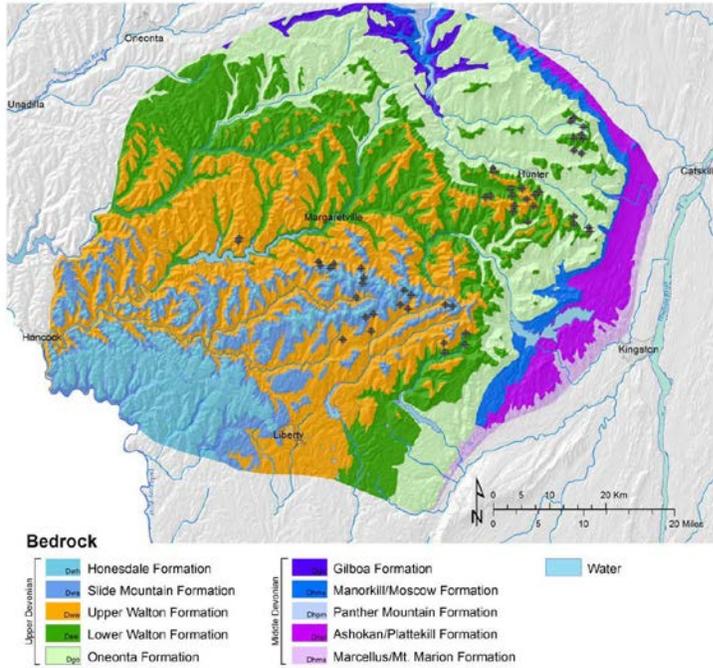


Figure 7. Soil samples were collected from within a 0.37-m² frame, allowing for the direct measurement of soil mass per unit land area



Table 1. Average soil nutrient pools in 50 forest soil profiles in the Catskills

Nutrient pools in forest vegetation at the Hubbard Brook Experimental Forest in 2007 are included for reference.

Adapted from Johnson, 2013

Horizon/ Layer	Soil Nitrogen (kg ha⁻¹)	Exch. Ca (kg ha⁻¹)	Exch. Mg (kg ha⁻¹)	Exch. K (kg ha⁻¹)
O _i +O _e	340	64	9	13
O _a /A	80	12	2	2
0-10 cm	1160	131	21	23
10-20 cm	800	71	12	15
20 cm – C Horizon	1530	125	19	26
Total	3910	403	63	80
Hubbard Brook Trees	792	710	69	298

Upland soils in the Catskills are relatively shallow, averaging 49.5 cm to bedrock or C horizon (Figure 8). They are also highly acidic, with average pH values ranging from 3.89 in the upper mineral soil layer to 4.75 in the O_i+O_e horizon (Figure 9). Organic horizons have higher concentrations of base cations (Ca, Mg, K, Na) than mineral soils (Figure 9), but because of low soil mass in the O horizons the pools of Ca, Mg, and K, which are essential plant nutrients, are small (Table 1).

Figure 8. Distribution of soil depth at 50 upland sites in the Catskills forest

Data from Johnson, 2013

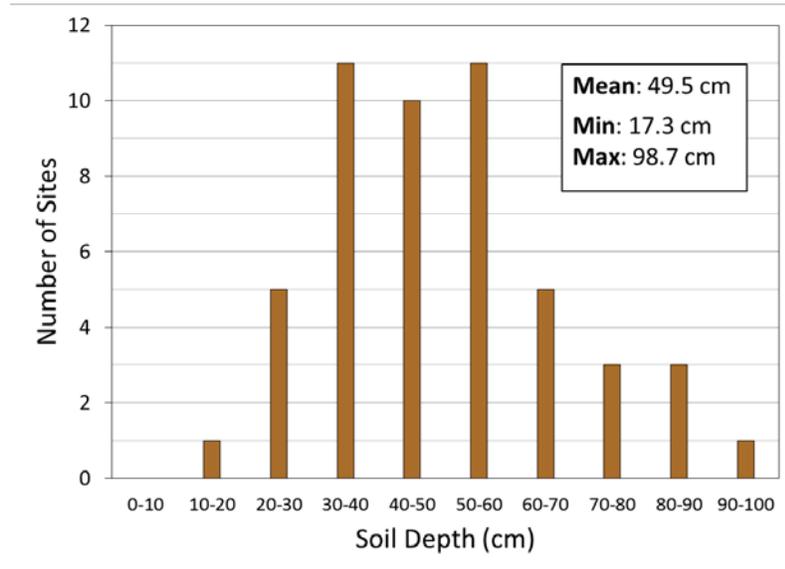
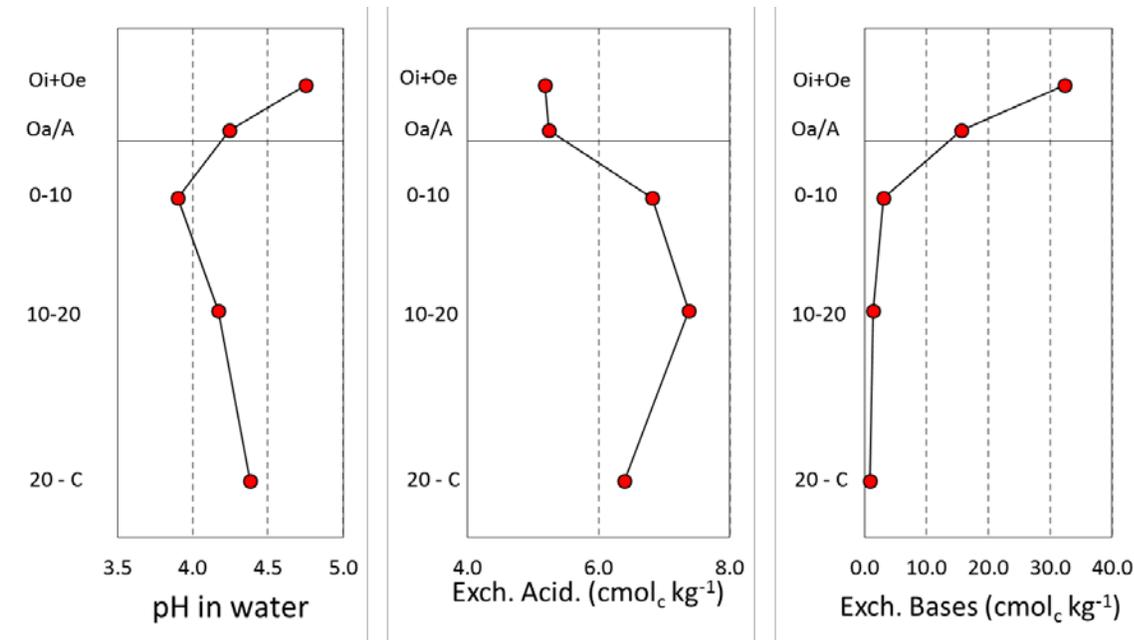


Figure 9. Depth profiles showing average pH, exchangeable acidity (in 1 M KCl), and exchangeable base cations (in 1 M NH₄Cl) for 50 forest soil profiles in the Catskills

Data from Johnson 2013



The data suggest that base cation pools in Catskills forest soils may be insufficient to sustain forest productivity and water quality services in the long term. The authors do not have estimates of the nutrient content of forest biomass in Catskills forests. However, a comparison with the Hubbard Brook Experimental Forest (HBEF) in New Hampshire, which has a similar species composition to much of the Catskills, suggests that pools of exchangeable Ca and K in Catskills soils are less than half of the amount in forest vegetation (Table 1). Similarly, Ca export in stream water is large relative to soil exchangeable pools. For example, stream water at Biscuit Brook in the southern Catskills, one of the watersheds in this study, carries 20 kg ha⁻¹ yr⁻¹ of Ca out of its watershed. This annual export represents 5% of the average exchangeable Ca pool in the soils studied (Table 1; Johnson 2013). These comparisons suggest that the historic depletion of exchangeable base cations from these soils, particularly Ca, may have significant implications for forest sustainability in the 21st century. Rebuilding base cation pools in these soils depends on the gradual weathering of minerals and, in some places, may require base cation amendments using lime or other mineral sources.

5 Total Maximum Daily Loads (TMDLs) in Adirondack Watersheds

Despite the large reductions in sulfur (S) and nitrogen (N) emissions in the United States, surface waters in the Adirondacks and Catskills are only slowly recovering. This observation raises two linked questions:

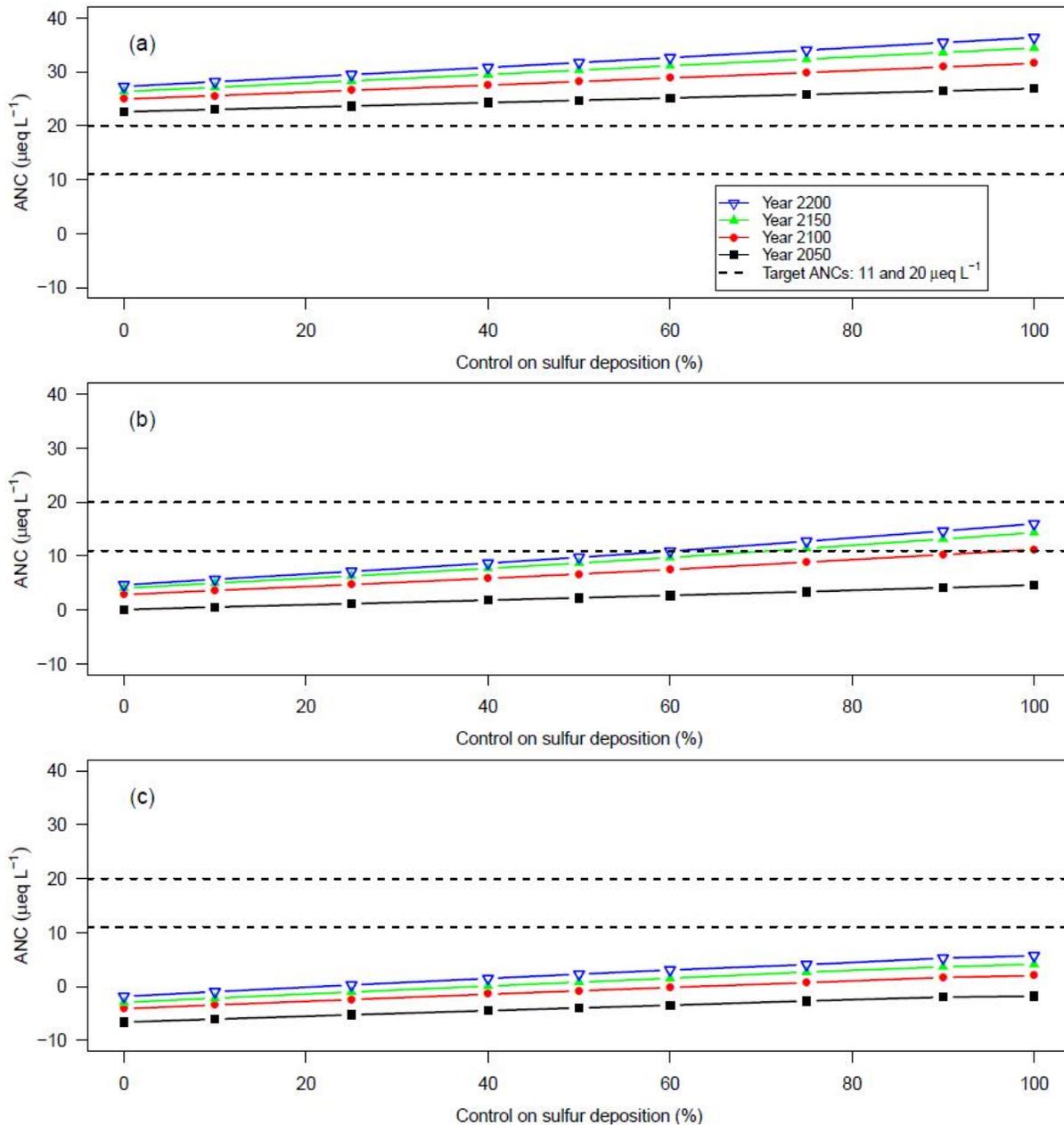
- What further reductions in atmospheric acid deposition are required for sensitive aquatic systems to attain a satisfactory water quality?
- Are there aquatic systems that are unlikely to ever attain a satisfactory level of water quality?

To address these questions, PNet-BGC, a comprehensive watershed-ecosystem model was used to simulate the response of 128 lake watersheds in the Adirondacks to various reductions in sulfur, nitrogen, and combined S+N emissions. From these simulations, a novel tool called the ANC response curve was developed. It can be used to display time-trends of ANC under different deposition reduction scenarios to determine total maximum daily loads (TMDLs).

Sets of ANC response curves are shown in Figure 10 for three lakes: Limekiln Lake (high-ANC), Constable Pond (low-ANC), and Peaked Mountain Lake (chronically acidic). The two threshold ANC values used – 11 and 20 $\mu\text{eq/L}$ – have been proposed as safe levels for brook trout, an important sport fish in the region. Each line on the response curve represents a different year and the intersection of a line with a target value shows the deposition reduction that would be required to attain the target value in that year. For example, in Constable Pond (Figure 10b), a 60% reduction in sulfur deposition would produce the 11 $\mu\text{eq/L}$ ANC target in the year 2200, while a 100% reduction in sulfur deposition would produce the same result one hundred years sooner.

Figure 10. Acid neutralizing capacity (ANC) response curves for three Adirondack lake watersheds

a) Limekiln Lake, a medium-till drainage lake; (b) Constable Pond; and (c) Peaked Mountain Lake, both thin-till drainage lakes (from Fakhraei et al. 2014). Each line shows the modeled lake-water ANC in a particular year under a range of sulfur emission reductions from 0-100%. Reference lines for ANC = 11 and 20 $\mu\text{eq/L}$ represent two estimates of minimum ANC to maintain brook trout.



Responses of Adirondack lake watersheds to reductions in atmospheric acid deposition are diverse (Figure 10). Limekiln Lake (Figure 10a), a medium-till drainage lake requires no further reductions in atmospheric sulfur deposition to maintain ANC concentrations above 20 $\mu\text{eq/L}$. Constable Pond (Figure 10b) and Peaked Mountain Lake (Figure 10c), both thin-till drainage lakes, are not likely to reach an ANC of 20 $\mu\text{eq/L}$ by the year 2200, even with 100% reductions in sulfur deposition. However, Constable Pond could reach the lower threshold of 11 $\mu\text{eq/L}$ if sulfur deposition were to be decreased by 60% or more. Our results indicate that some ponds, like Peaked Mountain Pond, are unlikely to recover a viable trout fishery even under the most aggressive control programs.

Based on these analyses, a TMDL for sulfur deposition is estimated to be 1.27 $\text{kg ha}^{-1} \text{yr}^{-1}$. This value, which includes a margin of safety, is a 66% reduction from the current average sulfur deposition in the Adirondacks. Therefore, approximately 68% of the lakes in the Adirondacks that currently have ANC below 20 $\mu\text{eq/L}$ are predicted to rise above 20 $\mu\text{eq/L}$ by 2150 if this TMDL were enforced.

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