

Regional Assessment of the Response of the Acid–Base Status of Lake Watersheds in the Adirondack Region of New York to Changes in Atmospheric Deposition Using PnET-BGC

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Understanding the response of soil and surface waters to changes in atmospheric deposition is critical for guiding future legislation on air pollution. The Adirondack region of New York experiences among the most severe ecological impacts from acidic deposition. The region is characterized by considerable variability in atmospheric deposition, surficial and bedrock geology, hydrologic flow paths, and vegetation resulting in variability in effects of acidic deposition. In this study, an integrated biogeochemical model (PnET-BGC) was applied to 37 forest lake watersheds to assess the response of soil and surface waters of the Adirondacks to changes in atmospheric deposition at a regional scale. Model-simulated surface water chemistry was validated against data from two synoptic surveys conducted in 1984 and 2001. Results indicate that the model is able to capture the observed changes in surface water chemistry during this period. The model was further used to forecast the response of soil and surface waters to three future emission control scenarios. Results indicate that under the Clean Air Act, surface water SO_4^{2-} concentrations will continue to decrease at a median rate of $-0.38 \mu\text{eq/L-yr}$, and surface water ANC is predicted to increase at a median rate of $0.11 \mu\text{eq/L-yr}$. More aggressive emission reductions will accelerate the rate of recovery. Under an aggressive control scenario, which represents an additional 75% reduction in SO_2 emissions beyond the implementation of the Clean Air Act, surface water SO_4^{2-} concentrations are predicted to decrease at a median rate of $-0.88 \mu\text{eq/L-yr}$, and surface water ANC is predicted to increase at a median rate of $0.43 \mu\text{eq/L-yr}$. Model predictions of several biologically relevant chemical indicators are also reported.

Introduction

Emissions of sulfur dioxide (SO_2) and nitrogen oxides (NO_x) from combustion of fossil fuels are transported long distances, potentially affecting forest and aquatic ecosystems over large geographic areas (1). Deposition of strong acids (e.g., HNO_3 , H_2SO_4) impacts forest health through both direct effects of precipitation and cloudwater on foliage and indirect effects of changes in soils that alter nutrient uptake by roots (1).

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Acidic deposition also acidifies surface waters, resulting in declines in species richness and decreases in abundance of zooplankton, macroinvertebrates, and fish (2).

As a result of the 1970 and 1990 Amendments to the Clean Air Act (CAAA), total SO_2 emissions nationwide have decreased approximately 30% below the 1980 levels (3). Wet sulfur (S) deposition has decreased more than 45% in the northeastern United States (3). In response to these decreases, surface waters in the Northeast including the Adirondacks have shown significant decline in sulfate (SO_4^{2-}) concentrations, with some showing increases in acid-neutralizing capacity (ANC; 3, 4). In contrast, NO_x emissions slightly increased from 1980 and 1990 and have remained constant since 1990 (1). Although a trend of decreasing surface water NO_3^- concentrations has been observed, this pattern does not reflect current patterns in NO_x emissions or wet nitrogen (N) deposition (3). Increasing NO_3^- leaching as a result of chronic high N loadings to forest ecosystems also contributes to soil and surface water acidification (5).

Of the regions affected by acidic deposition, the Adirondack region of New York has experienced among the most severe ecological impacts (6). In a survey of 1469 lakes ($>200 \text{ m}^2$ in surface area) during 1984–1986, 24% of the lakes were found to be fishless (6). Adirondack watersheds are also characterized by considerable variability in atmospheric deposition, hydrologic flow paths, surficial and bedrock geology, and vegetation composition, resulting in considerable variability in the sensitivity to acidic deposition. The response of complex ecosystems in the region to past, current, and future changes in atmospheric S and N deposition is of great interest. In this study, we quantified the response of soil and surface waters in the region to changes in atmospheric deposition at a regional scale through the application of an integrated biogeochemical model (PnET-BGC). The model was applied to 37 lake watersheds in the Adirondacks, providing the first assessment on the response of soil and surface waters across the entire region to effects of acidic deposition through the use of a comprehensive biogeochemical model. Although other acidification models (7, 8) have been applied to watersheds in the region (9, 10), our analysis was conducted at a regional scale relying on geographic information systems (GIS) to parameterize PnET-BGC, considers the simulation of both S and N transformations, and includes estimates of future emission changes.

Methods

Model Description. PnET-BGC is a comprehensive forest–soil–water model developed by linking a monthly C, N, and water balance model PnET (11) with a soil model BGC to allow for comprehensive simulations of element cycling within forest and the interconnected aquatic ecosystems (12). The model is able to simulate both abiotic processes and biotic processes. Especially the representation of biomass accumulation and the associated element cycling enable the evaluation of land disturbance and climatic events on soil and water chemistry (12). The model uses relatively simple formulations and requires a moderate number of inputs to quantify the acid–base status of soil and surface waters under various levels of atmospheric deposition; therefore, it can serve both as a research and a management tool. Its simplicity also makes it a good candidate for regional applications. Although other models (e.g., MAGIC, 7; ILWAS, 8; NuCM, 13; CHUM, 14; ETD, 15; Birkenes, 16) have been used to simulate effects of acidic deposition, these models either lack representation of biotic processes or are limited by large input requirements. A description of the PnET-BGC model includ-

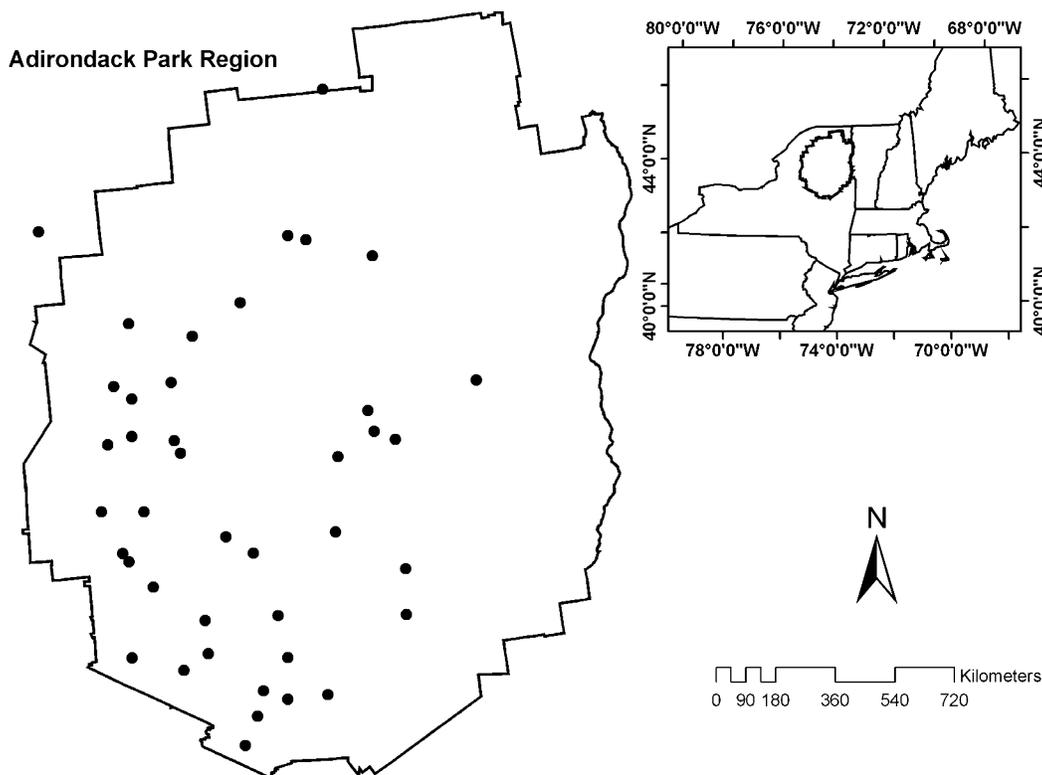


FIGURE 1. Locations of the Direct/Delayed Response Program lake watersheds in the Adirondack region of New York.

TABLE 1. Summary of the Characteristics of the Direct/Delayed Response Program Lake Watersheds in the Adirondack Region of New York (10)

variables	median	range
latitude (deg)	43.8	43.1–44.7
longitude (deg)	74.6	73.8–75.1
elevation (m)	565.7	418.5–791.0
watershed (ha)	206.7	36.3–1782.2
lake area (ha)	14.8	3.9–398.9
percent shallow deposit (%)	46.7	0–100
wetland percentage (%)	2.1	0–12.8
hardwood (%)	76.0	3.2–100
coniferous (%)	9.4	0–57.7
mixed forest (%)	21.1	0–96.1
wet S deposition (kg of S/ha-yr)	9.1	7.6–10.1

ing a detailed sensitivity analysis of parameter values is available in Gbondo-Tugbawa et al. (12).

Study Region. The 37 lake watersheds in the Adirondacks included in this study are among the 145 watersheds in the northeastern United States that were statistically selected by the Direct/Delayed Response Program (DDRP) initiated by U.S. EPA in 1984 to represent the acid-sensitive watersheds in the region (>4 ha in lake area; 9, 10; Figure 1). The watersheds selected receive a wide range of acidic deposition and exhibit considerable variability in elevation, watershed area, lake area, vegetation composition, and sensitivity to acidic deposition (Table 1). Among the 37 lakes, 28 lakes have ANC values less than 50 $\mu\text{eq/L}$. Vegetation of the region is dominated by northern hardwoods with red spruce and balsam fir at higher elevation and conifers adjacent to the lakes. Wetlands are a common landscape feature in the region, and many watersheds have extensive wetland coverage. Many forests in the Adirondacks have undergone severe disturbances including fire, logging, and hurricane impacts (6, 17, 18). These lake watersheds also show considerable variability in historical land disturbance. Nine of these 37 watersheds were largely burned during 1903–1908.

Model Inputs. Deposition and Climate. Regional regression models of wet S, N, base cation and Cl^- deposition, and dry S and N deposition as a function of latitude and longitude are available for the northeastern United States and were used to derive deposition inputs based on location of each site (19). The model was developed based on data from 26 wet and 13 dry deposition monitoring sites across the northeastern United States (19). PnET-BGC estimates dry deposition based on user inputs of dry to wet deposition ratios. Therefore, dry deposition estimated from the regression model was converted into dry to wet deposition ratios. Dry to wet deposition ratios for base cations and Cl^- were derived from the DDRP study, in which dry deposition was estimated based on the relationship between annual wet deposition and the ambient air concentration (9).

Using relationships between current emissions and deposition (20) plus estimates of past emissions (21), we reconstructed historical patterns of atmospheric wet deposition of S, N, and base cations at the Huntington Forest (HF) in the Adirondacks (22). In previous model applications, a constant dry to wet deposition ratio was assumed for each ion (12). However, based on data from several dry deposition monitoring sites in the Northeast, a relationship was found between air SO_2 concentrations and dry to wet S deposition ratios, suggesting decreases in dry to wet S deposition ratios in response to decreasing air SO_2 concentrations (23). This relationship along with the relationship between air SO_2 concentrations and SO_2 emissions were used to derive the historical changes in dry to wet S deposition ratios at the HF (23). The reconstructed temporal pattern was applied to all the sites as scalars. A throughfall study in a central Adirondack watershed suggested that S deposition under coniferous were 2.5 times greater than deciduous forests (24). Therefore the enhanced collection of dry S deposition under coniferous and mixed forests were accounted for by applying an enhancement factor (2.5 and 1.75 for coniferous and mixed forests, respectively) to the dry-to-wet ratios estimated from the regression models. Dry deposition for

TABLE 2. Summary of Target Emissions of SO₂ and Percent Reduction in SO₂ and NO_x Emissions under Three Future Scenarios: Base Case, Moderate Control, and Aggressive Control Scenarios Based on Current Proposals on Emission Reductions

scenarios	utility SO ₂ (million ton)	non-utility SO ₂ (million ton)	reduction in SO ₂ ^a (%)	reduction in NO _x ^a (%)
1990 CAAA (base case)	8.95	5.6	~40	~5
moderate control	6	5.6	~55	~20
aggressive control	1	5.6	~75	~30

^a Percent reduction is relative to the 1990 values and would be achieved by 2020.

each watershed was estimated as a weighted percentage of the area of deciduous, coniferous, and mixed forests.

Climate inputs were derived from regression models developed by Ito et al. (25) for the Adirondacks (i.e., precipitation, temperature) and solar radiation was derived from models developed by Aber and Freuder (26) for the northeastern United States.

Land Disturbance History. Past land disturbances have considerable impact on N cycling within forest ecosystems (27, 28). Site-specific land disturbance history was generally derived from maps from the Adirondack Park Agency (APA) and the descriptions of McMartin (18) and Sullivan et al. (17). For sites that experienced logging prior to 1890, 20% biomass mortality was assumed. Logging after 1920 was considered to be slight. Therefore for sites that experienced logging after 1920, we assumed 20% cutting at a 20-yr rotation. A severe hurricane in 1950 impacted many forests and was followed by salvage logging (18). An APA map was used to derive hurricane-affected areas for each watershed for use in model simulations. We assumed that 40% of the downed biomass was removed by salvage in the affected areas.

Soil Parameters. Soil parameters of soil mass, cation exchange capacity (CEC), and cation exchange coefficients were derived from the 1984 DDRP survey data (9, 10). Soil pH-dependent SO₄²⁻ adsorption parameters were derived from experimental data from Woods Lake (Driscoll, C. T., unpublished) as described in Gbondo-Tugbawa et al. (29).

Vegetation Type. PnET models (PnET-CN and PnET-BGC) use generalized inputs of four different vegetation types including northern hardwoods, spruce-fir, red maple-red oak mixture, and pine. For model calculations, vegetation species identified in DDRP were reclassified into these types and parameters for the dominant type at each watershed were used in model calculations.

Wetland Retention. To account for the retention of SO₄²⁻ and NO₃⁻ within wetlands, an algorithm for simulating S and N retention within lakes developed by Kelly et al. (30) was adopted for use in PnET-BGC. The retention coefficient is calculated as a function of a mass transfer coefficient, annual runoff, and wetland percentage within the watershed.

Future Scenarios. Current emission reduction proposals call for 50–75% reduction in utility emissions of SO₂ beyond full implementation of Title IV of the 1990 CAAA and 70–75% reduction from 1997 levels in NO_x emissions by various dates (1). To evaluate the effects of possible future emission controls, we considered three future control scenarios: (i) the base case (1990 CAAA); (ii) a moderate control scenario; and (iii) an aggressive control scenario (Table 2). The moderate control scenario considers emission reductions somewhat less than the most modest proposed reduction, and the aggressive control scenario considers emission reduction of somewhat more aggressive than the most aggressive reduction proposed. Therefore, these two

scenarios bound the range of emission reductions called by various proposals. The emission reductions are assumed to be implemented through linear decreases from 2010 to 2020.

Results

Model Simulated Surface Water Chemistry in 1984 and 2001. Chemistry data of these DDRP lakes are available through a synoptic survey conducted by the U.S. EPA during the fall of 1984 after lake turnover (9, 10) and another survey conducted by Warby et al. (31) during summer 2001. In our study, chemistry data from the 1984 survey were used to parameterize model inputs of mineral weathering rates for supply of base cations. The chemistry data in 2001 were mainly used to validate the ability of the model to simulate temporal changes in lake chemistry from 1984 to 2001. It was assumed that these index surveys are indicative of the annual volume weighted concentrations. Determining the rates of change in lake chemistry during 1984–2001 using data from these two synoptic surveys may not be adequate, given large inter-annual variations that can occur. Therefore, data from the Adirondack Long Term Monitoring (ALTM; 4) program were used to supplement our analysis of temporal changes in lake chemistry. Data from the ALTM program are based on monthly measurements of lake chemistry during 1982–2001 at 16 lakes.

Model-simulated lake concentrations of SO₄²⁻, Ca²⁺, and Mg²⁺ in 1984 generally agreed with the observed data (Figure 2a–c). Model simulated surface water pH and ANC also agreed well with survey data for both 1984 and 2001 ($R^2 = 0.62–0.84$; Figure 2d,e), although the model seems to underpredict pH for values >5.5. Model simulated annual concentrations of NO₃⁻ for 1984 ranged between 1 and 29 $\mu\text{mol/L}$. Surface water NO₃⁻ concentrations in the northeastern United States generally show large seasonal variability, with the lowest NO₃⁻ concentrations occurring during growing season as a result of plant uptake and denitrification, and highest concentrations occurring during snowmelt (32). Thus NO₃⁻ concentrations obtained from fall or summer surveys are not comparable with the simulated annual volume-weighted concentrations. Simulated annual NO₃⁻ concentrations increase 2.1 $\mu\text{mol/L}$ during 1984–2001. However the ALTM data show decrease in lake NO₃⁻ concentrations of 3.9 $\mu\text{mol/L}$ during 1982–2000, although many (67%) of the lakes do not have statistically significant trends (4).

Simulated lake SO₄²⁻ concentrations in 2001 generally captured the mean and the extent of variation in the observed SO₄²⁻ concentrations (Figure 2a). However due to the relatively narrow range exhibited in SO₄²⁻ concentrations, correlation between simulated and observed concentrations was relatively low. Nonetheless, simulated median rate of change in lake SO₄²⁻ during the period of 1984–2001 compared well with the DDRP lake surveys and the long-term trend in the ALTM lakes (Figure 3a). The simulated rates of change in lake SO₄²⁻ are relatively uniform, similar to the patterns observed in the ALTM lakes. However, changes exhibited in DDRP lake surveys showed larger variability (Figure 3a).

Model simulated concentrations of Ca²⁺ and Mg²⁺ in 2001 also showed larger discrepancies from the observed data ($R^2 = 0.72, 0.51$, Figure 3b). The model predicted less decrease in Ca²⁺ concentrations than observed in the survey data, thus overpredicting Ca²⁺ in 2001. Model-simulated average rates of decreases in Ca²⁺ were also smaller than changes observed in the ALTM lakes. This discrepancy could be partially attributed to the decreases in NO₃⁻ concentrations that were not captured by the model. Overall, model-simulated rates of change in Ca²⁺+Mg²⁺ were fairly uniform across the region, and agree well with trends observed in the

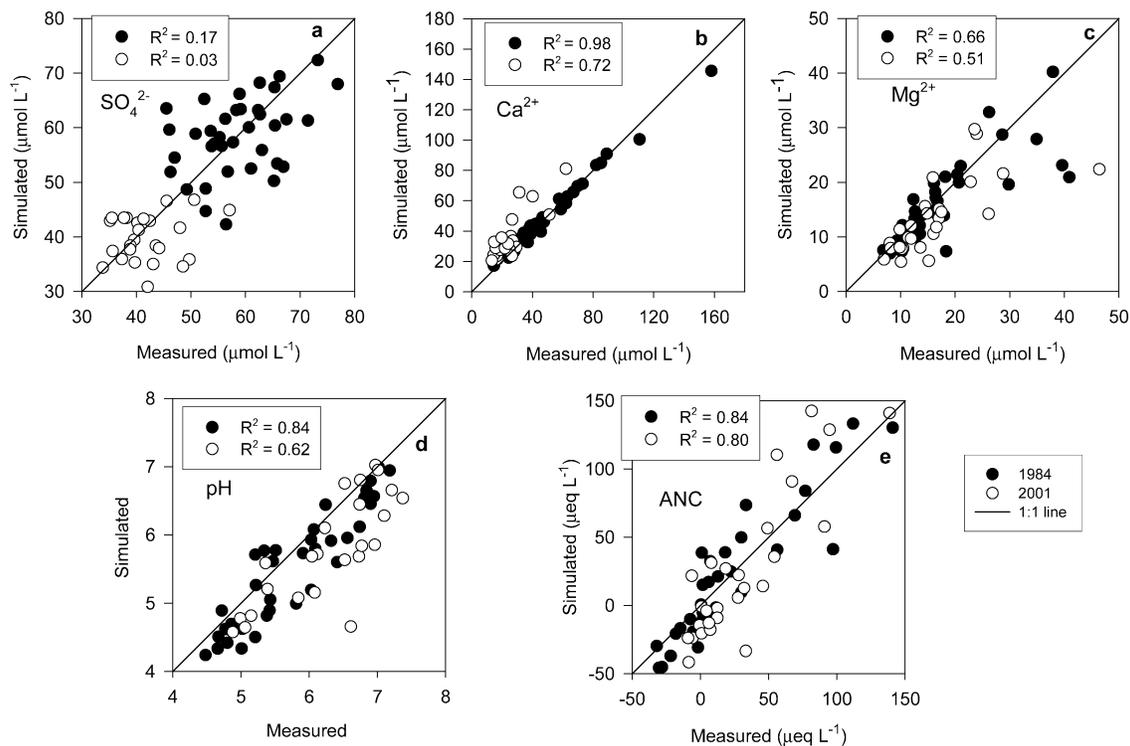


FIGURE 2. Model-simulated annual volume-weighted concentrations of SO_4^{2-} (a), Ca^{2+} (b), and Mg^{2+} (c), pH (d), and ANC (e) in 1984 (in black circle) and 2001 (in open circle), as compared with synoptic survey data in 1984 conducted by the U.S. EPA (10) and 2001 conducted by Warby et al. (25).

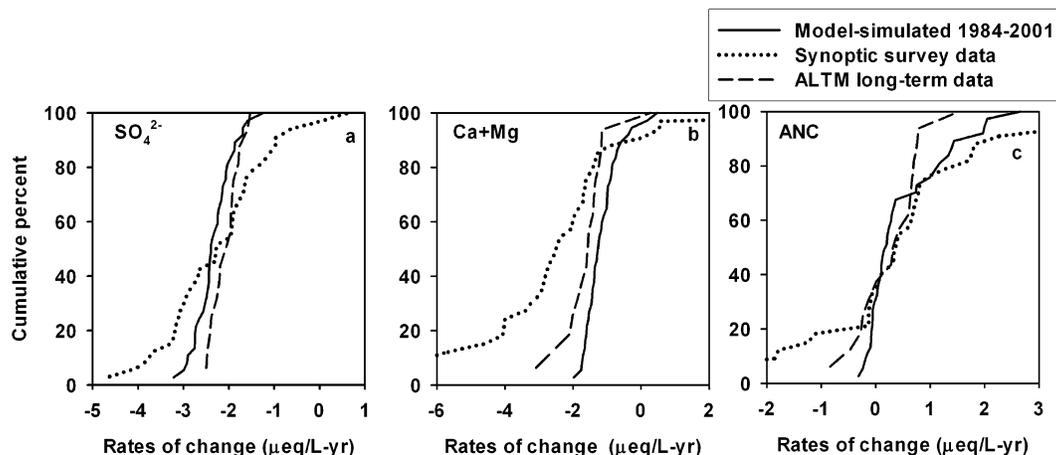


FIGURE 3. Model simulated rates of change in lake SO_4^{2-} (a), $\text{Ca}^{2+} + \text{Mg}^{2+}$ (b), and ANC (c) during 1984–2001 as compared with estimates from synoptic surveys in 1984 and 2001. The rates of change from synoptic surveys were estimated as difference in concentrations of 1984 and 2001 divided by 17 yr. Long-term trends during 1982–2000 determined based on monthly data from the 16 Adirondack Long-Term Monitoring Program lakes were also included for comparison.

ALTM lakes. In contrast, larger variability was found in the survey data (Figure 3b).

Model-predicted ranges of changes in ANC were comparable to changes observed both in the DDRP lake surveys and the ALTM lakes (Figure 3c). Predicted rates of change in ANC are also relatively uniform compared to the survey data. Both the ALTM data and the survey data indicate approximately 35% of the lakes exhibit continuing acidification (i.e., ANC decreases). Model predictions generally agree well with these observations.

Historical Acidification. Model hindcast of pre-anthropogenic (~1850) conditions indicate acidic deposition has greatly altered soil and surface waters in the region. Simulated pre-anthropogenic conditions showed low concentrations of SO_4^{2-} , NO_3^- , and Al_i and high pH, ANC, and soil %BS compared to the current values (i.e., 1984, Table 3).

TABLE 3. Model Simulated Pre-anthropogenic Lake Chemistry and Soil %BS (median values, $\mu\text{eq/L}$, except pH and %BS) as Compared to 1984 Survey Values

	preanthropogenic	1984
SO_4^{2-}	9.6	116.6
NO_3^-	4.8	0–35
pH	6.4	5.8
ANC	85.6	7.8
Al_i	0.3	0.7
%BS	20.8	9.4

Response to Future Scenarios. Model simulations indicate that in response to the three future control scenarios, during 2010–2050, SO_4^{2-} concentrations at these DDRP lakes decrease at rates ranging from -0.3 to -0.7 , -0.5 to -0.9 ,

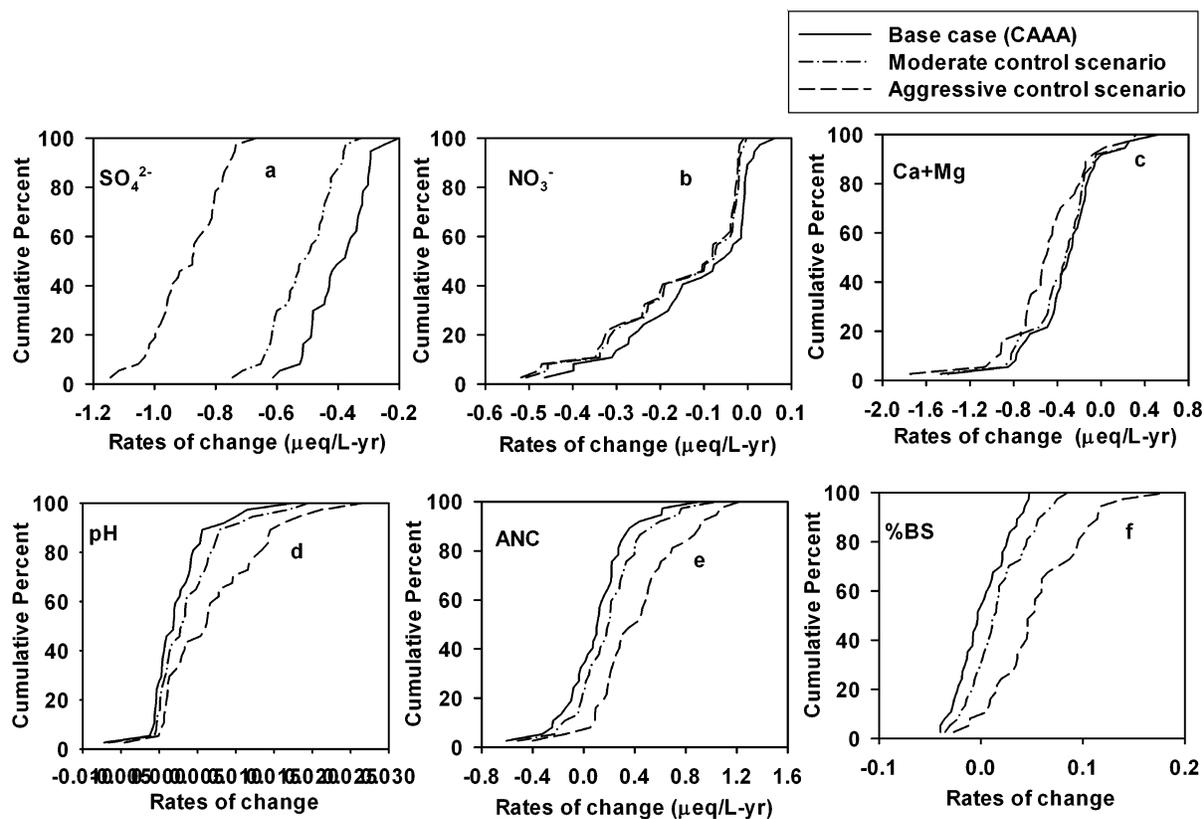


FIGURE 4. Model simulated rates of change in lake SO_4^{2-} (a), NO_3^- (b), $\text{Ca}^{2+} + \text{Mg}^{2+}$ (c), pH (d), ANC (e), and soil %BS (f) during 2010–2050 under base case, the moderate control and the aggressive control scenarios.

and -0.9 to $-1.3 \mu\text{eq/L-yr}$ under the base case, the moderate control, and the aggressive control scenarios, respectively (Figure 4a). The moderate and aggressive control scenarios accelerate the median rates of decreases in SO_4^{2-} by 0.2 and $0.5 \mu\text{eq/L-yr}$, respectively. Predicted rates of change in SO_4^{2-} for the period of 2010–2050 are lower than the rates of change currently observed in the ALTM lakes (mean $-2.27 \mu\text{eq/L-yr}$ during 1982–2000; 4). The three scenarios yielded distinctly different lake SO_4^{2-} concentrations at steady state (i.e., 2050), with the aggressive scenario resulting in the lowest SO_4^{2-} concentrations with values ranging between 20 and $40 \mu\text{eq/L}$ (Figure 5a).

Predicted lake NO_3^- concentrations under the three scenarios generally show slight decreases (-0.5 to $0.05 \mu\text{eq/L-yr}$), with moderate and aggressive controls slightly enhancing the rates of NO_3^- decrease (Figure 4b). Predicted NO_3^- concentrations in 2050 under the moderate and aggressive control scenarios are also slightly lower than the base case (Figure 5b).

Predicted lake $\text{Ca}^{2+} + \text{Mg}^{2+}$ concentrations decrease at most of the sites (85%) (Figure 4c), in response to the predicted decreases in SO_4^{2-} and NO_3^- concentrations. The predicted rates of decrease in base cations are lower than rates of SO_4^{2-} and NO_3^- decrease, resulting in predictions of increasing pH and ANC at most sites (Figure 4d,e).

Under the three future scenarios, predicted lake pH generally increase at rates ranging from 0 to 0.03 unit/yr, with $<10\%$ of the sites showing decreases in pH. Rates of pH increase are also enhanced under the moderate and aggressive control scenarios. Similarly, predicted lake ANC during 2010–2050 increases at most of the sites (0 to $1.2 \mu\text{eq/L-yr}$; Figure 4e), with moderate and aggressive control scenarios accelerating the median rates of ANC increase (0.20 and $0.43 \mu\text{eq/L-yr}$ vs $0.11 \mu\text{eq/L-yr}$ under base case). Model predicted rates of ANC increase under the aggressive scenario are generally lower than rates currently observed

in the ALTM lakes (mean rate of $0.78 \mu\text{eq/L-yr}$ for the 7 out of 16 ALTM lakes that showed significant ANC increase during 1982–2000; 4). In a previous study, PnET-BGC was applied to four intensively studied lake-watersheds in the Adirondacks (33). Predicted rates of ANC increase for these four study sites (0.22 – $0.37 \mu\text{eq/L-yr}$) correspond to the 30–50 percentiles of the rates predicted for the DDRP lakes. Predicted soil percent base saturation (%BS) increases at a median rate of $0.05\%/yr$ under the aggressive control scenario, with 20% of the sites showing continuing decreases (Figure 4 f).

Discussion

The model application to these regional sites indicated the difficulty in capturing the full variation exhibited in lake SO_4^{2-} concentrations (Figure 2a). The departure is probably due to the high variability in S deposition across complex terrain (e.g., elevation, aspect, vegetation composition, edges; 34, 35), which is not represented by the empirical deposition model (19), coupled with the use of generalized algorithms that could not fully depict the spatial variations in watershed S dynamics. For northeastern United States, wetland retention has been found to be an important factor regulating watershed retention of SO_4^{2-} (9, 10, 23). The current model uses a simplified relationship to describe wetland S retention. Future work should focus on developing more accurate estimates of atmospheric deposition based on landscape features and quantifying the influence of wetlands in watershed S retention in order to improve model simulations. Research could also be done to evaluate effects of forest type, soil adsorption, and hydrologic flow paths on watershed S retention.

A trend of decreasing in NO_3^- concentrations has been recently observed in the ALTM lakes and other surface waters in the northeastern United States (3, 4) despite constant emissions of NO_x and atmospheric wet NO_3^- and NH_4^+

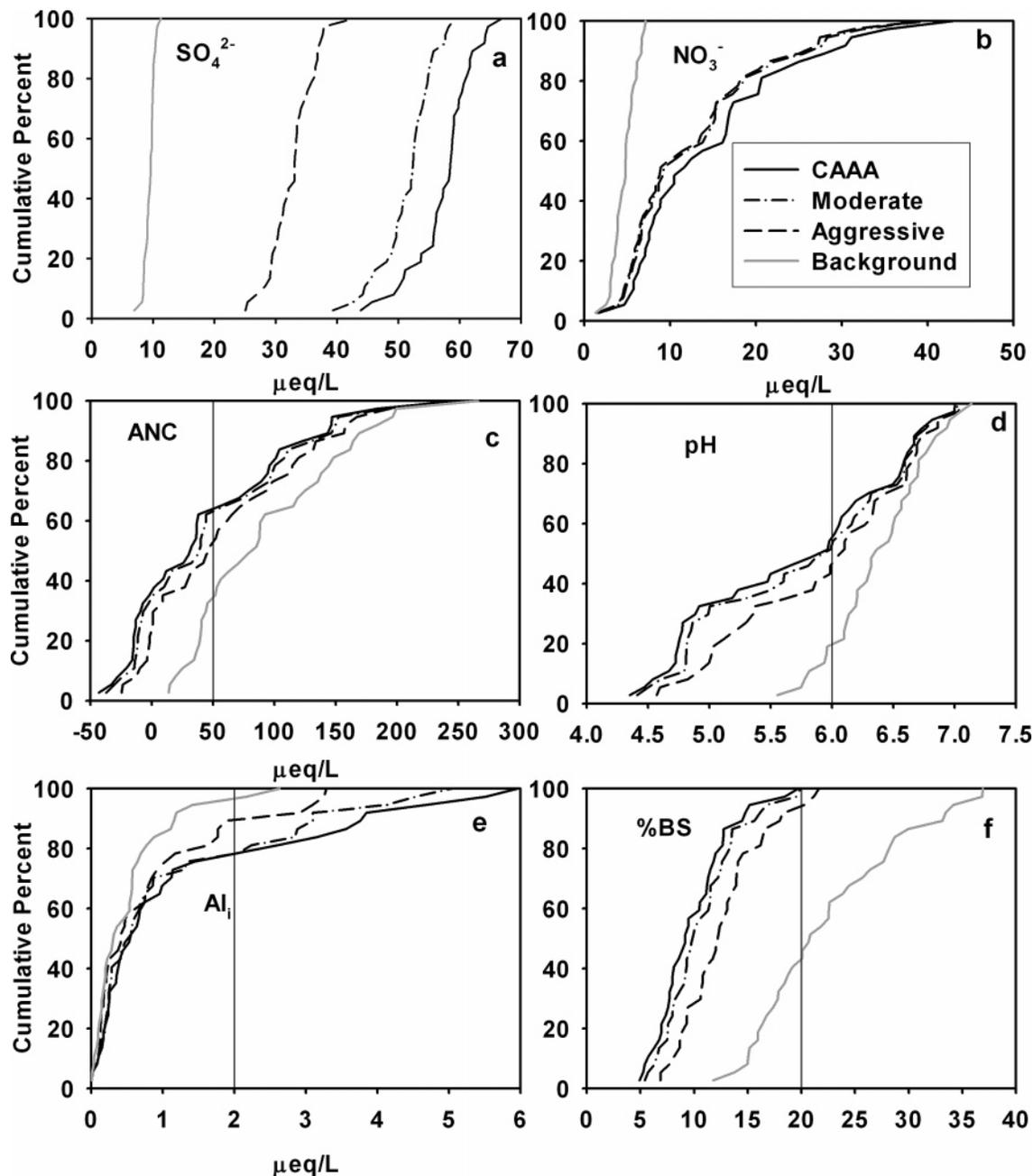


FIGURE 5. Model simulated concentrations in lake SO_4^{2-} (a), NO_3^- (b), Al_i (c), pH (d), ANC (e), Ca/Al ratio (f), and %BS (g) in soil in 2050 under the base case, the moderate control, and the aggressive control scenarios.

deposition. Model calculations, however, predict increase in NO_3^- . For watersheds in the northeastern United States that receive chronic elevated N loadings, both N saturation theory (36) and nutrient retention theory (37) would predict increases in NO_3^- leaching. As a result, the observed decrease in NO_3^- is unexpected, and climate variations are speculated as the most plausible cause (38). However, the exact mechanism(s) for this response is still not known. Research is needed to investigate the possible causes and mechanisms for the increases in watershed N retention.

Recovery of ecosystems from stress associated with acidic deposition following control of acidifying emissions is beneficial changes in chemical conditions that will improve the environmental quality for forest and aquatic biota. Specifically, chemical recovery should result in improvement in some biologically relevant indicators above the threshold values: the molar Ca/Al ratio of soil water > 1.0 and soil BS > 20% (for the health of forest vegetation; 39, 40), and surface

water pH > 6.0, ANC > 50 $\mu\text{eq/L}$; and decrease in inorganic monomeric Al concentrations < 2 $\mu\text{mol/L}$ (for health of aquatic biota; 41).

Despite the marked improvements in surface water pH and ANC under the aggressive control scenario, a large percentage of lakes (~40–50%) are still predicted to have pH and ANC values below threshold values in 2050 (Figure 5c, d), and around 10% of the lakes still have Al_i greater than 2 $\mu\text{mol/L}$ (Figure 5e). While most of the lake watersheds have soil %BS below the critical value of 20% in 2050 (Figure 5f), predicted Ca/Al ratio in soil solutions are generally above threshold value of 1.

Model hindcasts of pre-anthropogenic conditions however suggest some of these lake watersheds may be naturally acidic and recovery to the threshold values of biologically relevant chemical indicators might not be achieved. Simulations indicate around 35% lakes have background ANC < 50 $\mu\text{eq/L}$, and 45% of the lake watersheds have background BS

< 20%. While most of lakes have pH >6 (80%) and Ali concentrations < 2 $\mu\text{mol/L}$ (95%).

Little is known about biological recovery, but it is likely biological recovery will occur only if chemical recovery is sufficient to allow the survival and reproduction of biota, and it is likely to occur years or decades after chemical recovery is achieved (1). Research has shown some aquatic biota (e.g., zooplankton) are likely to recover approximately 10 years after chemical condition is restored (42). Given the chemical recovery predicted for these Adirondacks lakes, full biological recovery may not be achieved by 2050.

Model sensitivity analysis to parameters by Gbondo-Tugbawa et al. (12) showed that model predictions of ANC are most responsive to variability in soil CEC, soil mass, and DOC adsorption coefficient. Sensitivity of ANC to soil CEC and soil mass are 0.35 and 0.23, respectively, indicating a 10% change in the parameter values will result in 3.5% and 2.3% change in predictions of ANC. Predictions of soil percent base saturation are most responsive to selectivity coefficients of Ca^{2+} and Al^{3+} (sensitivity of 2.33 and 1.81) and partial pressure of CO_2 in the soil (sensitivity of 3.20). Soil CEC and selectivity coefficients of Ca^{2+} and Al^{3+} were derived from the DDRP database, in which soils were sampled based on 39 classes, and site-specific soil parameters were derived through aggregation of the 39 sampling classes (8, 9). Therefore, the estimates of these parameters should be representative of the soils in these lake watersheds. Sensitivity of ANC to soil mass is relatively small. For the DOC adsorption coefficient and the partial pressure of CO_2 , we used the values reported by Gbondo-Tugbawa et al. (12).

The model predictions were based on the assumption constant climate over the period of 2010–2050. Given the marked influence of climate on surface water chemistry especially for NO_3^- (43), model predictions are aimed at projecting long-term trends instead of inter-annual variations. Model predictions were also conducted under the assumptions of constant base cation, NH_4^+ and Cl^- deposition, mineral weathering, and no additional land disturbance during 2010–2050. Deviations from these assumptions could alter model projections.

This study is the first application of PnET-BGC at the regional scale. The investigation has both research and management implications. The application tested the ability of the model to reproduce spatial and temporal patterns in surface water chemistry. We also assessed the response of a full range of soils and surface waters in the region to changes in atmospheric deposition. Model calculations indicate that emission reductions generally result in beneficial changes in soil and surface water chemistry. Recovery is predicted to be widespread; however, complete chemical and biological recovery will not be achieved by 2050 even under the most aggressive emission control scenarios considered.

Acknowledgments

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