

REGIONAL APPLICATION OF AN INTEGRATED BIOGEOCHEMICAL MODEL TO NORTHERN NEW ENGLAND AND MAINE

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Abstract. An integrated biogeochemical model (PnET-BGC) was applied to 60 Direct/Delayed Response Program (DDRP) lake watersheds in northern New England and Maine to investigate processes regulating spatial and temporal patterns in lake SO_4^{2-} concentrations, past changes in the acid–base status of soil and surface waters, and their response to future emission control scenarios. Model simulations indicate that watershed elevation is an important factor influencing inputs of atmospheric S deposition to the watersheds and thus spatial patterns in lake SO_4^{2-} concentrations. Wetland S retention, vegetation composition, and thickness of surficial deposits were also found to influence lake SO_4^{2-} concentrations. Model simulations also suggest that decreases in lake SO_4^{2-} concentrations observed during 1984–2001 were mostly a result of decreases in atmospheric S deposition. Decreases in lake SO_4^{2-} concentrations were coupled with a near-stoichiometric decline in base cation concentrations. Comparison of atmospheric deposition of base cations and simulated drainage loss during this period indicates that decreases in atmospheric base cation deposition were partially responsible for decreases in lake base cation concentrations. Decreases in drainage loss of base cations were mainly a result of declines in net loss from soil exchange sites associated with decreases in SO_4^{2-} leaching. Model forecasts generally suggest further decreases in SO_4^{2-} concentrations and increases in acid neutralizing capacity (ANC) in lake water and soil percent base saturation (BS) at most of the DDRP lake watersheds under three future emission control scenarios, with the more aggressive control scenarios resulting in faster recovery from acidic deposition. Under an aggressive control scenario, surface water ANC is simulated to increase at a median rate of $0.30 \mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$ in northern New England and $0.22 \mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$ in Maine. However, although marked improvement is expected, recovery to background conditions will probably not occur before 2050.

Key words: acidic deposition; acid neutralizing capacity; base cations; Clean Air Act; modeling; recovery; sulfate.

INTRODUCTION

Concerns over acidic deposition and its widespread impacts on terrestrial and aquatic ecosystems across Europe and North America have led to national and international legislation reducing emissions of acid forming substances (SO_2 and NO_x ; Stoddard et al. 1999). Across the northeastern United States, concerns over the health of red spruce and sugar maple, and decreases in species richness of fish have been reported since the early 1950s. The Title IV of the 1990 Amendments of the Clean Air Act (CAAA) was established to reduce effects of acidic deposition by reducing emissions of SO_2 from electric utilities (Driscoll et al. 2001). As a result of the 1970 and 1990 CAAs, surface waters in northeastern United States have shown widespread decreases in SO_4^{2-} concentrations, and increases in acid neutralizing capacity (ANC) have been reported in some surface waters (Driscoll et al. 2003, Stoddard et al. 2003).

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Northern New England (NNE) and Maine (ME) are among the subregions of the United States that have been impacted by acidic deposition. Many forests, soils, and surface waters in these two subregions are sensitive to acidic deposition. A recent assessment of Long-Term Monitoring (LTM) trends in surface water chemistry indicated that SO_4^{2-} concentrations in New England region have declined, coupled by a stoichiometric decline in base cation concentrations and no significant change in ANC (Stoddard et al. 2003). Forests and aquatic biota of these subregions thus are still under the stress from acidic deposition. The objectives of this study were to investigate factors contributing to these trends in surface water chemistry and to determine whether acidification stress will continue under several future emission scenarios, exploring the extent of recovery that might occur over the next 50 years.

We conducted a regional application of an integrated biogeochemical model (PnET-BGC) at 60 sites in these two subregions in order to accomplish these objectives. The model PnET-BGC was first developed and applied to a northern hardwood forest, the Hubbard Brook Experiment Forest (HBEF) in New Hampshire (Gbondo-

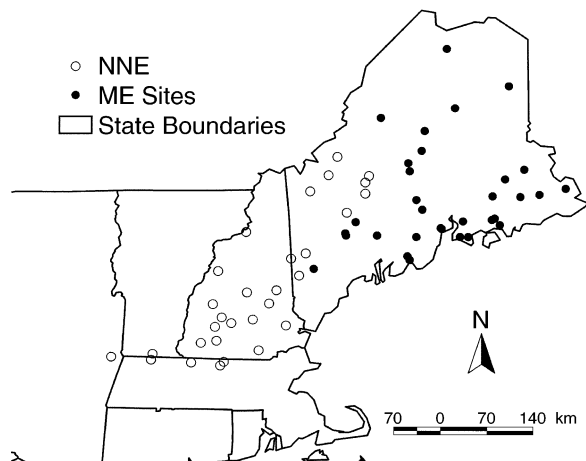


FIG. 1. Locations of Direct/Delayed Response Program (DDRP) sites in Northern New England (NNE) and Maine (ME) subregions.

Tugbawa et al. 2001). The model links a C, N and water model (PnET; Aber et al. 1997) with a soil biogeochemical model (BGC; Gbondo-Tugbawa et al. 2001) to simulate cycling of major elements within northern forests and reactions in soil and surface waters. The 60 lake watersheds located in NNE and ME were statistically selected by the Direct/Delayed Response Program (DDRP) initiated by the U.S. EPA to represent the acid-sensitive watersheds in the northeastern United States (Fig. 1; Church et al. 1989). Chemistry of these lakes was surveyed during National Surface Water Survey in 1984 (Linthurst et al. 1986, Landers et al. 1988). To evaluate the response of these lakes to recent changes in atmospheric deposition, these lakes were resurveyed in the summer of 2001 and results from these two surveys during different time periods were compared (R. A. F. Warby, C. E. Johnson, and C. T. Driscoll, *unpublished manuscript*). Here, we present a synthetic analysis of the results from the model application to these lake watersheds, including S dynamics, N status, changes in base cations, and surface water ANC, as well as other critical indicators in response to past emissions and future control scenarios.

S DYNAMICS

Base-case simulation

Atmospheric S deposition.—Across the northeastern United States, wet S deposition generally decreases from the west to the east, with dry deposition decreasing from more urban areas of the south to the north (Ollinger et al. 1993). Total S deposition thus shows a spatial pattern of decreasing values from southwest to northeast. This gradient in deposition was found to generally regulate the spatial pattern in lake SO_4^{2-} concentrations across the northeastern United States (Church et al. 1989, Driscoll et al. 1998). To reflect this gradient, we calculated average deposition for each

watershed for the period 1980–1990 by multiplying SO_4^{2-} concentrations in precipitation by precipitation quantity estimated from regional empirical models of Ollinger et al. (1993, 1995). Ollinger et al. (1993) estimated SO_4^{2-} concentrations in precipitation as a function of latitude, while precipitation was estimated as a function of latitude, longitude and elevation. The latitude, longitude, and elevation of the lake were used to estimate wet deposition for the entire lake watershed.

Since the mid 1800s, there has been considerable change in SO_2 emissions in the United States (Driscoll et al. 2001). Wet S deposition has followed this trend, as indicated by a close relationship between annual SO_2 emissions for the source area of the Northeast and annual SO_4^{2-} concentrations in bulk deposition at the HBEF (Likens et al. 2001). This relationship along with the reconstruction of historical emission records has been used to derive the temporal changes in wet S deposition at the HBEF for the period 1850–1963 (Gbondo-Tugbawa et al. 2002). The reconstructed deposition reflects trends in annual SO_4^{2-} concentrations. However, monthly variations in precipitation quantity were accounted for by averaging long-term monthly precipitation. For 1963–1998, the direct measurements of monthly SO_4^{2-} concentrations in precipitation and monthly precipitation quantity were used to derive the monthly bulk S deposition (data available online).² In order to apply the reconstructed temporal patterns in bulk S deposition to all the sites in NNE, these values were converted into scalars by standardizing to the annual average deposition of 1980–1990 (from measurements) at HBEF. Temporal patterns of wet S deposition for sites in NNE therefore were estimated by multiplying these scalars with the annual average deposition of 1980–1990 estimated from models of Ollinger et al. 1993. Similarly for sites in Maine, temporal patterns in wet deposition were first reconstructed based on data from a National Atmospheric Deposition Program (NADP) site located in the Bridgton, Maine (ME02; data available online)³ and applied to all the sites as scalars.

In previous model applications, the dry-to-wet S deposition ratio was assumed to be constant and dry S deposition was assumed to change in the same proportion relative to wet deposition (e.g., Gbondo-Tugbawa et al. 2002). Data from Clean Air Status and Trends Network (CASTNET; data available online)⁴ and Atmospheric Integrated Research Monitoring Network (AIRMoN; data available online)⁵ suggest a close relationship between dry-to-wet S deposition ratios and air SO_2 concentrations across the northeastern United States, indicating dry-to-wet deposition ratios could vary with SO_2 concentrations in the air (Chen and Driscoll

² <http://www.hubbardbrook.org/research/data/data.htm>

³ <http://nadp.sws.uiuc.edu/sites/>

⁴ <http://www.epa.gov/castnet>

⁵ <http://www.arl.noaa.gov/research/programs/airmon.html>

coll 2004). Therefore, dry-to-wet deposition ratios may vary over time. At HBEF, relative changes in dry S deposition appear to be different than bulk deposition (Palmer et al. 2004). The observed relationship between air SO₂ concentrations and dry-to-wet deposition ratios as well as relationship between air SO₂ concentrations and SO₂ emissions were used to reconstruct the temporal patterns in dry S deposition. The reconstructed temporal patterns in dry S deposition were again converted into scalars and applied to all the DDRP sites in the NNE and ME subregions.

SO₄²⁻ adsorption coefficients.—In the PnET-BGC model, a pH-dependent SO₄²⁻ adsorption isotherm was used to simulate adsorption/desorption of SO₄²⁻ within soil (Gbondo-Tugbawa et al. 2001, 2002). The DDRP study provided detailed characterization of vegetation, geology, soil properties (e.g., adsorbed SO₄²⁻ concentrations, soil pH, exchangeable cation concentrations) and other watershed and lake characteristics (e.g., watershed area, lake residence time, wetland area) for each lake watershed (Church et al. 1989). Soil properties for each watershed were based on aggregating 38 soil sampling classes in the region. In our analysis, these estimates of soil properties were used to derive model inputs. The estimates of exchangeable cation concentrations were used to derive cation exchange selectivity coefficients. The estimates of adsorbed SO₄²⁻ in soil, soil pH and lake SO₄²⁻ concentrations (as a surrogate of soil solution SO₄²⁻ concentrations) were used to derive the pH-dependent adsorption coefficients for the soils in watersheds of the NNE and ME subregions, according to the approach described by Gbondo-Tugbawa et al. (2002).

Wetland retention.—Although rates of S reduction within wetlands could vary with locations within the watershed and portion of watershed drainage through wetlands, watershed S retention was found to increase with the percentage of wetlands in the DDRP watersheds (Church et al. 1989). The relationship developed by Kelly et al. (1987) to calculate in-lake retention of S thus was adopted to simulate S retention within wetlands:

$$R_s = \frac{S_s}{(R_o/P_w) + S_s} \quad (1)$$

where R_s is the retention coefficient of S in wetlands, S_s is the mass transfer coefficient (m/yr), R_o is the runoff (m/yr), and P_w is the wetland percentage within the watershed. An assumed mass transfer coefficient of 2 m/yr seems to yield good agreement between simulated S retention and observed data (Chen and Driscoll 2004). Thus, for our base case simulation, Eq. 1 and this mass transfer coefficient were used to calculate S retention within wetlands.

Residual analysis

The simulated lake SO₄²⁻ concentrations in 1984 showed little variation when compared to the 1984 sur-

vey data. As indicated in a previous study (Chen and Driscoll 2004), the lack of variation in predictions could be attributed to (1) variations in inputs of atmospheric deposition that are not fully captured by the regional regression models; (2) watershed characteristics (e.g., soil depth) are not well characterized in the available data sets and/or; (3) biogeochemical processes (e.g., wetland retention) that contribute to variations in lake SO₄²⁻ that are not well represented in the model.

We attempted to improve model predictions by first examining the relationship between the residuals in model predictions and a series of watershed characteristics, including geographic position, wetland coverage, vegetation composition, and surficial geology, which were available from the DDRP study (Church et al. 1989). We then identified the watershed characteristic that was most responsible for the residuals and attempted to address the apparent bias in predictions by either formulating a simple algorithm in the model to better represent that process or modifying model inputs. We then reexamined the residuals and repeated the above steps until no significant relationships were found between the residuals and the available watershed characteristics in the DDRP data set. Although not part of the model simulation, limitations in the available data sets and the regional nature of the application suggested that this analysis was critical in order to better quantify the variations in regional lake SO₄²⁻ concentrations.

For residual analysis, we evaluated the effectiveness of simulations using two criteria: the root mean square error (RMSE) and efficiency (Eff). The RMSE and Eff are defined as

$$\text{RMSE} = \sqrt{\sum_{i=1}^n (\hat{Y}_i - Y_i)^2 / n} \quad (2)$$

$$\text{Eff} = \frac{\sum_{i=1}^n (\hat{Y}_i - \bar{Y}_i)^2}{\sum_{i=1}^n (Y_i - \bar{Y}_i)^2} \quad (3)$$

where \hat{Y}_i and Y_i are the predicted and measured value for site i , n is the total number of sites, and \bar{Y}_i is the average measured value from all sites. Smaller RMSE values represent less error resulting from model simulations. An Eff of 1 indicates the model is able to reproduce all the variance in the measured data. Initial simulations for lake SO₄²⁻ in the northern New England subregion had a RMSE of 19.2 μeq/L and Eff of 0.50. RMSE and Eff for the Maine subregion were 20.3 μeq/L and 0.63, respectively.

For sites in both NNE and ME, elevation was found to be the most significant watershed characteristic that contributed to the residuals (Table 1). In both subregions, significant negative correlations between the residuals and elevation indicated that the influence of

TABLE 1. Correlations between residuals and watershed characteristics for base-case simulations and simulations with modifications in watershed characteristics and RMSE and Eff values for the simulations.

Parameter	Subregion							
	NNE simulations				ME simulations			
	1	2	3	4	1	2	3	4
Latitude	-0.15	-0.12	-0.12	-0.15	-0.45	-0.09	-0.16	-0.08
Longitude	-0.15	0.05	0.09	0.11	0.05	0.18	0.18	0.07
Elevation	-0.59	0.15	0.25	0.21	-0.61	-0.14	-0.22	-0.15
Wetland (%)	-0.03	0.02	-0.08	-0.10	0.38	0.47	0.31	0.23
Wetland area	0.08	-0.32	-0.31	-0.22	0.44	0.39	0.21	0.22
Wet vegetation	-0.05	-0.03	0.00	-0.02	0.32	0.30	0.20	0.12
DOC	-0.16	0.06	-0.06	-0.07	0.33	0.49	0.28	0.25
Depth to bedrock	-0.25	-0.23	-0.23	-0.23	0.43	0.40	0.34	0.34
BRD_LT2†	-0.26	-0.33	-0.21	-0.14	-0.22	-0.11	-0.12	-0.23
BRD_LT3‡	-0.36	-0.30	-0.15	-0.16	-0.13	-0.13	-0.16	-0.23
Mean flow path	0.19	0.05	0.18	0.20	0.17	0.11	0.04	0.11
Coniferous (%)	-0.11	0.12	-0.03	-0.05	-0.02	-0.06	0.10	0.10
Deciduous (%)	0.16	0.26	0.29	0.10	-0.20	-0.15	-0.14	-0.22
Mixed forest (%)	-0.17	-0.27	-0.32	-0.13	0.11	0.12	0.12	0.22
Pine (%)	0.02	0.01	0.04	0.08	0.40	0.33	0.36	0.32
Hardwood-spruce mixture	0.03	0.03	0.09	0.15	-0.52	-0.35	-0.47	-0.03
RMSE	19.2	15.9	15.0	13.4	20.3	16.6	12.5	10.1
Eff	0.50	0.77	0.97	0.90	0.63	0.54	0.66	0.67

Notes: Simulation 1 is the base-case simulation. Simulation 2 is corrected for errors caused by elevation. Simulation 3 is corrected for soil depth at NNE (northern New England) and wetland percentage at ME (Maine). Simulation 4 is corrected for mixed forest. The largest correlation for each simulation is shown in bold.

† BRD_LT2 is the percentage of watershed with depth to bedrock less than 0.5 m.

‡ BRD_LT3 is the percentage of watershed with depth to bedrock less than 1 m.

elevation on SO_4^{2-} concentrations was not negligible (Table 1; Fig. 2). For ME sites, the correlation between the residuals and latitude was also significant. This correlation was believed to be a result of high elevation coinciding with high latitude, as suggested by the correlation between elevation and latitude. In an analysis of SO_4^{2-} budgets for DDRP sites in Maine, Norton et al. (1988) observed that the amount of lake SO_4^{2-} unexplained by wet deposition decreased from northwest to southeast. They attributed the unexplained SO_4^{2-} to higher dry S deposition at northern high elevation sites. Wet S deposition was also believed to increase with elevation. Either way, the influence of elevation on lake SO_4^{2-} was probably manifested through inputs of atmospheric S deposition to the watersheds. We addressed this consideration by adding an elevation component into the SO_4^{2-} concentration model of Ollinger et al. (1993) in deriving the deposition inputs. The coefficient for the elevation term was specified to be the same as the model of Ito et al. (2002), assuming effects of elevation on deposition was the same in NNE and ME as the Adirondack region of New York. The original empirical model for average SO_4^{2-} concentration in precipitation during 1980–1990 was

$$\text{SO}_4^{2-} \text{ (in } \mu\text{eq/L)} = -599.19 + 10.08 \times \text{longitude}$$

where longitude is measured in degrees. Adding elevation to the model for the NNE sites became

$$\begin{aligned} \text{SO}_4^{2-} = & -599.19 + 10.08 \times \text{longitude} \\ & + 0.14(\text{elevation} - 300) \end{aligned}$$

where elevation is measured in meters. The adjusted

model for ME differed slightly for sites with elevation >115 m:

$$\begin{aligned} \text{SO}_4^{2-} = & -599.19 + 10.08 \times \text{longitude} \\ & + 0.14(\text{elevation} - 115). \end{aligned}$$

With consideration of elevation in wet deposition, RMSE for NNE sites decreased to $15.9 \mu\text{eq/L}$ and Eff increased to 0.77. RMSE for ME sites decreased to $16.6 \mu\text{eq/L}$ while Eff slightly decreased to 0.54.

For NNE sites, the percentage of shallow surficial deposits was the second most significant watershed characteristic that contributed to the residuals (Table 1). The model tended to underpredict lake SO_4^{2-} at watersheds with a larger percentage of shallow surficial deposits and overpredict SO_4^{2-} at watersheds with a smaller percentage of shallow deposits (Fig. 2). This discrepancy could be due to the relatively uniform soil depth we used in model simulations. There was no good characterization of soil depth at these DDRP watersheds except depth to bedrock; therefore we choose to use a relatively uniform soil depth of 0.5 m. For watersheds with a larger percentage of shallow surficial deposits, precipitation could be routed to surface waters by shallow flow paths limiting contact with the soil, resulting in higher lake SO_4^{2-} concentrations and vice versa. We tried to correct for this factor by specifying a shallower soil depth for sites with shallow surficial deposits (BRD_LT3 $> 40\%$; where BRD_LT3 = percentage of watershed with depth to bedrock less than 1 m) and using depth to bedrock as soil depth for sites with little area of shallow surficial deposits (BRD_LT3

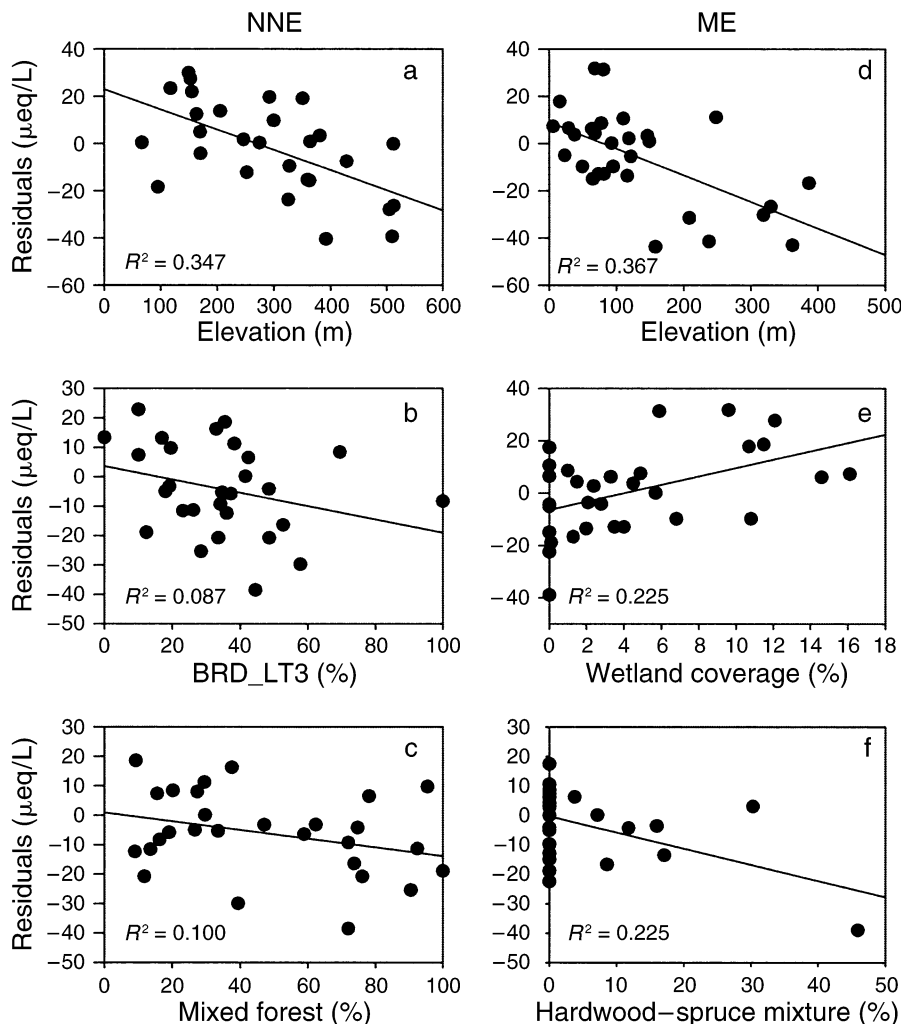


FIG. 2. Relationships between residuals and the watershed characteristics for NNE sites (a–c) and ME sites (d–f). BRD_LT3 is the percentage of watershed with depth to bedrock less than 1 m.

< 20%). Using a shallower soil depth did not improve model simulations at watersheds with shallow surficial deposits, indicating the underprediction at these sites is probably due to other factors. The use of a deeper soil depth for watersheds with thick surficial deposits, however, significantly improved model results. Thus we corrected for the influence of thickness of surficial deposits by changing soil depth to depth to bedrock at sites with thick surficial deposits (BRD_LT3 < 20%). The resulting simulations reduced RMSE to 15.0 µeq/L and improved Eff to 0.97.

With the correction of elevation and surficial geology, vegetation emerged as another significant watershed characteristic for the NNE sites, showing negative correlations between residuals and the percentage of mixed forests (Fig. 2). Enhanced collection of dry S deposition in coniferous and mixed forests has been reported at many locations in the northeastern United States (e.g., Mollitor and Raynal 1982, Cronan 1985).

To correct for this effect, we used a dry deposition enhancement factor of 1.75 for mixed forests and 2.5 for coniferous forests based on Mollitor and Raynal (1982). Thus the new dry-to-wet deposition ratio could be estimated based on the original ratio, the percentage of deciduous forest, coniferous and mixed forest, and the enhancement factors. The resulting simulations significantly reduced RMSE, while Eff remained at 0.90.

For ME sites, wetlands appeared to be another significant watershed factor affecting lake SO_4^{2-} . The consistent positive correlation between residuals and wetland percentage, wetland area, and lake dissolved organic carbon (DOC) concentrations indicated that overprediction in lake SO_4^{2-} occurred at watersheds with a high wetland percentage (Table 1; Fig. 2). This pattern suggested that the assumed mass transfer coefficient of 2 m/yr was likely too low to effectively predict wetland S retention for sites in Maine. We accounted for this observation by specifying a larger mass transfer co-

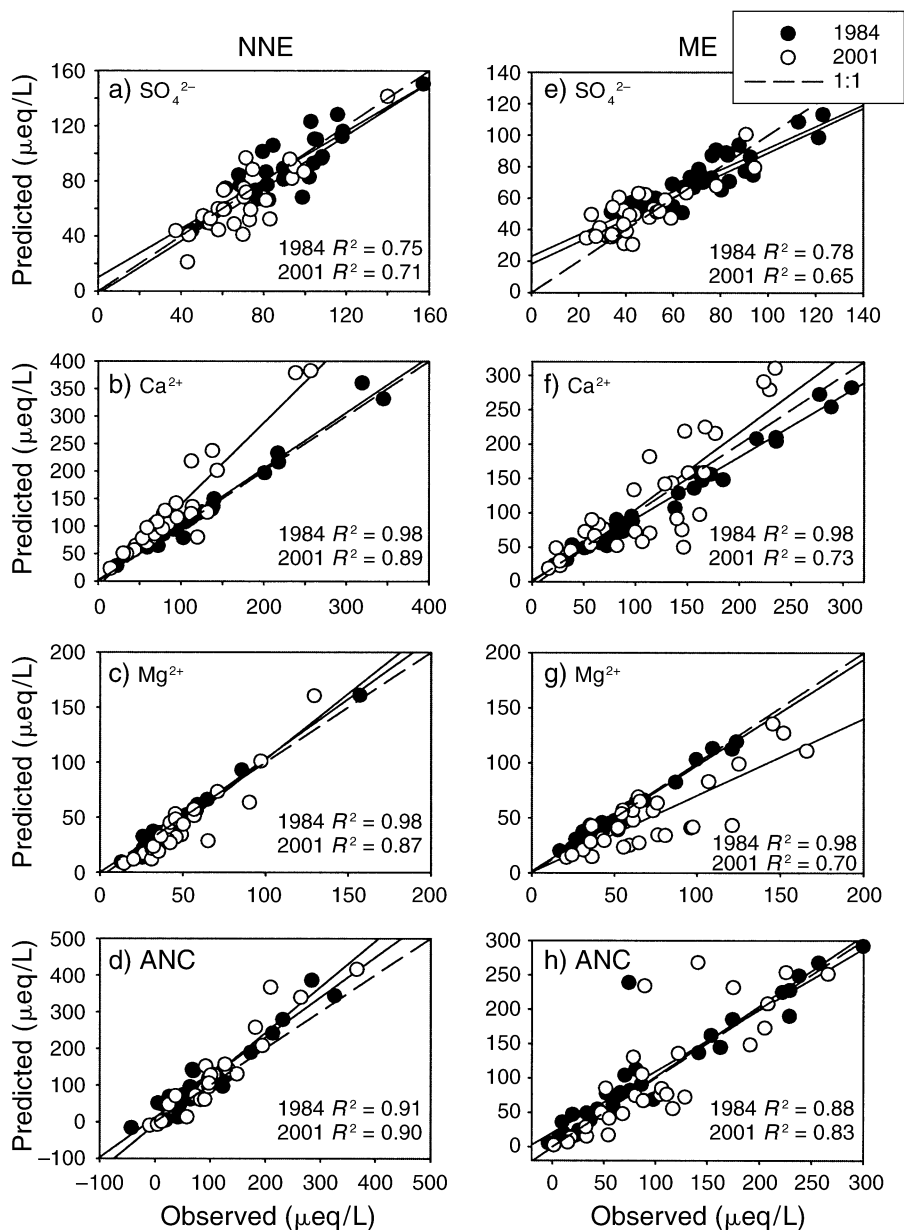


FIG. 3. Predicted concentrations of SO_4^{2-} , Ca^{2+} , Mg^{2+} , and ANC in 1984 (solid circles) and 2001 (open circles) compared to concentrations obtained from surveys for (a–d) NNE lakes and (e–h) ME lakes.

efficient for lakes with large wetland coverage in the watershed (>4%) and had overpredictions of lake SO_4^{2-} . The resulting simulations significantly decreased RMSE, but decreased Eff slightly (Table 1).

For sites in Maine, the residuals were found to be positively correlated with the percentage of pine forest coverage in the watershed and negatively correlated with the percentage of hardwood–spruce mixture forests, suggesting an overprediction in lake SO_4^{2-} under pine forests and an underprediction under mixed hardwood and spruce forests. It is not clear why the model overpredicted lake SO_4^{2-} in watersheds with pine for-

ests. However, mixed hardwood and spruce forests could influence watershed S dynamics through enhanced collection of dry S deposition. Thus similar to NNE sites, a dry deposition enhancements factor of 1.75 was applied to sites with mixed hardwood–spruce coverage. This correction further reduced the RMSE of the simulations to $10.1 \mu\text{eq/L}$.

By considering these factors, the model was able to explain about 75–78% of the variation in lake SO_4^{2-} concentrations (Fig. 3). The predicted spatial pattern of lake SO_4^{2-} generally agreed with the pattern exhibited in the 1984 survey.

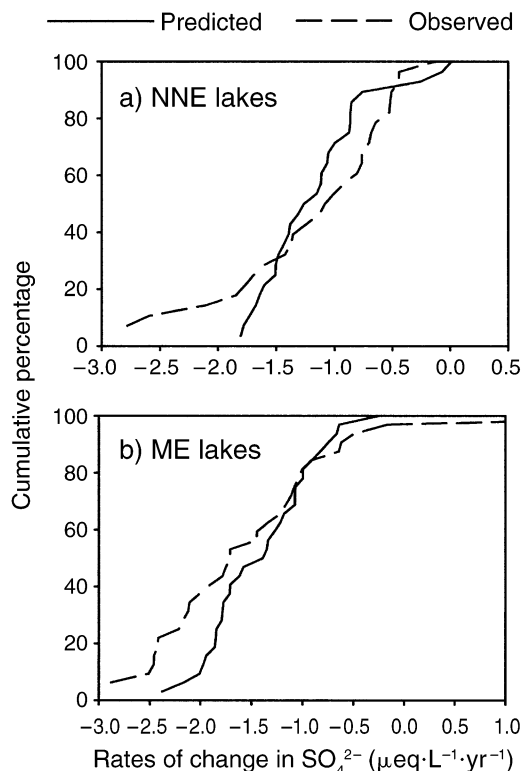


FIG. 4. Distribution of predicted rates of change in SO_4^{2-} compared to rates derived from surveys for (a) NNE lakes and (b) ME lakes for 1984–2001.

Changes in SO_4^{2-} concentrations in response to changes in atmospheric deposition

In response to declines in atmospheric S deposition in recent years, the model predicted decreases in lake SO_4^{2-} concentrations in both subregions. The predicted lake SO_4^{2-} concentrations in 2001 compared well with concentrations measured in the survey during summer 2001 (regression $R^2 = 0.71$ for NNE sites and 0.65 for ME sites; Fig. 3). The predicted median rates of change in lake SO_4^{2-} of $-1.80 \mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$ for NNE and $-1.74 \mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$ for ME during 1984–2001 compared well with rates observed at long-term monitoring (LTM) sites in New England ($-1.77 \mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$; Stoddard et al. 2003). The predicted rates of change for 1984–2001 compared well with rates derived from the 1984 and 2001 synoptic surveys (Fig. 4).

The widespread decreases in lake SO_4^{2-} concentrations were mostly a result of decreases in S deposition resulting from SO_2 emission controls. A comparison of decreases in atmospheric S deposition and declines in drainage loss of SO_4^{2-} to surface waters indicated that changes in lake SO_4^{2-} were largely driven by changes in S deposition (Fig. 5a). Moreover the rates of decrease in drainage loss of SO_4^{2-} were closely related to rates of decrease in atmospheric S deposition (Fig. 5b). Predicted rates of change during 1990–2001 were greater than rates for the longer period of 1984–2001

(median rate of -1.80 and $-1.74 \mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$ vs. -1.21 and $-1.37 \mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$ for NNE and ME, respectively). The greater rates of change in lake SO_4^{2-} during 1990–2001 especially during 1996–2001 were mostly due to the additional SO_2 emission controls associated with the implementation of Title IV of the 1990 CAAA (Stoddard et al. 2003, Driscoll et al. 2003).

The estimated decreases in S deposition during 1984–2001 were $-0.026 \pm 0.007 \text{ g S}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$ for NNE and $-0.019 \pm 0.004 \text{ g S}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$ for ME. These rates of change in S deposition if fully translated into surface waters, would result in decreases in lake SO_4^{2-} of $-2.17 \pm 0.40 \mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$ for NNE and $-1.61 \pm 0.40 \mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$ for ME through mass balance calculations. However, predicted changes in lake SO_4^{2-} flux were $-0.015 \pm 0.005 \text{ g S}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$ for NNE sites and $-0.011 \pm 0.007 \text{ g S}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$ for ME sites. Predicted changes in lake SO_4^{2-} concentrations were $-1.16 \pm 0.48 \mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$ for NNE sites and $-1.40 \pm 0.51 \mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$ for ME sites. Model calculations suggested that the delayed response of surface water SO_4^{2-} was largely due to the net desorption of previous adsorbed SO_4^{2-} in response to decreases in atmospheric deposition (Fig. 5). For period of 1984–2001, predicted

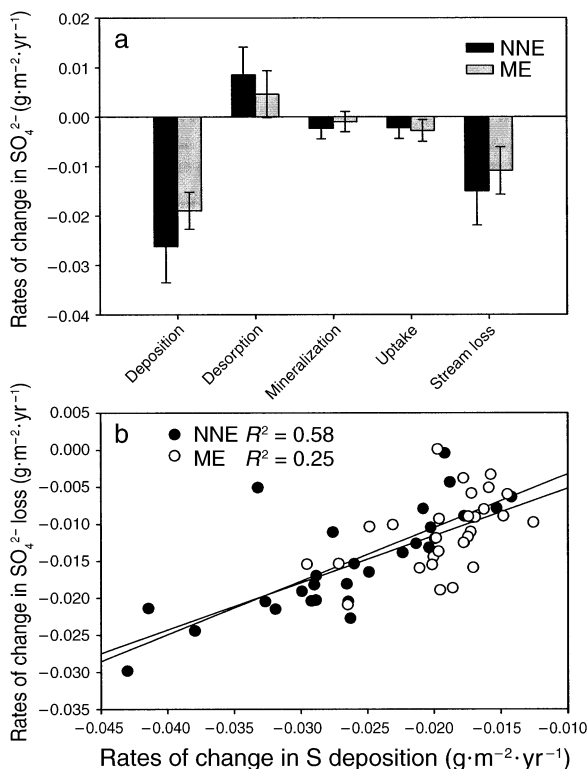


FIG. 5. (a) Estimated rates of change in atmospheric SO_4^{2-} deposition in relation to simulated changes in desorption, mineralization, uptake, and stream loss of SO_4^{2-} and (b) the relationship between changes in S deposition and lake watershed loss of SO_4^{2-} for Northern New England (NNE) and Maine (ME) over the period 1984–2001. Values in (a) are means \pm SD.

average desorption rates were $0.028 \pm 0.030 \text{ g S}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$ for NNE sites and $0.038 \pm 0.020 \text{ g S}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$ for ME sites. Note that predicted rates of net SO_4^{2-} desorption exhibited an increasing trend of $0.009 \pm 0.006 \text{ g S}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$ for NNE and $0.005 \pm 0.005 \text{ g S}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$ for ME during this period. The rates of net SO_4^{2-} desorption agreed with the rates previously estimated for the HBEF by Gbondo-Tugbawa et al. (2002). Model predictions indicate smaller effects of mineralization and vegetation uptake on S retention during this relatively short period (Fig. 5). The model predicted slight decreases in net S mineralization. These fluxes were almost completely balanced by decreases in net plant uptake of S (Fig. 5).

N DYNAMICS

Model inputs related to N dynamics

N deposition.—Similar to S deposition, N deposition also exhibits a distinct gradient across the northeastern United States, decreasing from southwest to northeast (Ollinger et al. 1993). There have also been considerable changes in NO_x emission and N deposition over the last 100 years. However unlike S, NO_x emissions and N deposition have remained relatively constant during the last 20 years. Similar to S, the spatial pattern of atmospheric N deposition was derived from ion concentrations in precipitation and precipitation quantity estimated from regional models of Ollinger et al. (1993, 1995). The temporal pattern of N deposition was also scaled from the reconstructed time series at the representative NADP site in each subregion.

Vegetation type.—Vegetation type has a strong influence on N dynamics in forested ecosystems (e.g., Aber et al. 2003, Lovett et al. 2002). In our model simulations, inputs for the dominant vegetation type were obtained from the original DDRP data (Church et al. 1989).

Land-disturbance history.—Past land disturbances have considerable impact on N cycling within forest ecosystems (Aber et al. 1997, Goodale et al. 2000). Forests of New England and Maine have experienced severe disturbances and generally followed a pattern of decreases in forest area through the late 1700s, a peak in agricultural lands from 1830–1890, and rapid reforestation from farmland abandonment through the late 19th and early 20th centuries (Foster 1995). A major hurricane in 1938 impacted forests in central and northern New England. Site-specific land-disturbance history is difficult to obtain for our study sites. For these watersheds, two major classes of disturbance patterns were assumed: (1) continuous forest (primary forests that have never been cleared), and (2) post agriculture (clearing for agriculture and reforestation on the abandoned field; Goodale et al. 2002). A large percentage of sites in New England and Maine contain open lands (pasture or abandoned farmland) and agricultural lands, suggesting possible agriculture activ-

ities in the past. Thus for sites with open lands or agriculture lands, we assumed the post agriculture land use pattern. While for sites without open land or agriculture land, continuous forest land use pattern was assumed. After Goodale et al. (2002), we assumed forest clearing in 1750 followed by continuous removal of 5% biomass through 1880 for post-agriculture land use and an 80-yr harvest cycling for continuous forest land use.

Simulated NO_3^- pattern

Predicted NO_3^- concentrations in 2001 were $3.66 \pm 2.38 \text{ }\mu\text{eq/L}$ at NNE sites and $1.81 \pm 1.12 \text{ }\mu\text{eq/L}$ at ME sites. These ranges of concentrations compared well with concentrations reported by Aber et al. (2003) for the region. The predicted rates of change in NO_3^- during 1984 and 2001 were generally small. During 1984 and 2001, the estimated average NO_3^- deposition was $0.45 \text{ g}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$ at NNE sites and $0.35 \text{ g}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$ at ME sites. The predicted NO_3^- leaching to surface water was around 0.05 and $0.03 \text{ g}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$ for NNE and ME, respectively, indicating strong N retention in these watersheds. The relatively large N retention at these sites was probably a result of extensive land disturbance in the past. Note that atmospheric N deposition and watershed NO_3^- leaching were lower in NNE and ME than in the Adirondack and Catskill regions of New York immediately to the west that exhibit higher NO_3^- leaching (Aber et al. 2003, Chen et al. 2004).

BASE CATIONS

Model inputs related to base cations

Base cation deposition.—Unlike S and N deposition, base cation deposition decreases from the coast to inland and from southeast to northwest (Ollinger et al. 1993). The regional models of base cation concentrations in precipitation chemistry and precipitation quantity as a function of geographic position by Ollinger et al. (1993) were used to derive wet deposition of base cations at each site. Again temporal patterns from long-term precipitation chemistry sites were scaled out for other sites.

Measurements or estimates of dry deposition of base cations are rare. Dry-to-wet deposition ratios estimated during the DDRP study for Na^+ , Mg^{2+} , K^+ , and Ca^{2+} were 1.61 ± 0.42 , 2.43 ± 0.53 , 2.24 ± 0.53 , and 1.09 ± 0.35 , respectively, for NNE and 1.12 ± 0.44 , 1.57 ± 0.65 , 1.98 ± 0.66 , and 0.81 ± 0.52 , respectively, for ME sites (Church et al. 1989). The estimated ratios for Na^+ and Mg^{2+} were higher than values summarized by Baker (1991; 0.37 and 0.75) for the northeastern United States. The ratios for K^+ and Ca^{2+} were very close to the values of Baker (1991; 2.21 and 1.19, respectively). In the Integrated Forest Study (IFS), Johnson (1992) found that dry-to-wet deposition ratios were generally <1 for Na^+ at sites without marine influence, <1 for Mg^{2+} except for one site in Maine, 1.2–3.3 for K^+ , and

<1 for Ca^{2+} at low-elevation eastern sites. Based on these values, the estimated dry-to-wet deposition ratios of K^+ and Ca^{2+} used for the DDRP were considered to be reasonable, while the ratios of Mg^{2+} might be overestimated. Given that watersheds in NNE and ME might be impacted by marine aerosols, the DDRP values of Mg^{2+} were still used in the model simulations.

Sea salt correction.—The inputs of base cations from marine salt spray were calculated based on a Cl^- mass balance. For sites with a Cl^- output that exceeded the estimated deposition, the unexplained portion of Cl^- in surface water was assumed to originate from either road salt or marine salt. We based our assumption of the Cl^- source for individual watersheds on their location (i.e., near coastal areas, adjacent to roads). Based on the estimated input of Cl^- from sea salt, inputs of other elements were calculated using their molar ratios to Cl^- in seawater (S/Cl, 0.10; Mg/Cl, 0.195; Ca/Cl, 0.038; K/Cl, 0.018; Na/Cl, 0.859). Inputs from sea salt were assumed to be constant over time.

Mineral weathering.—Direct measurements of weathering rates are not available for these sites, thus weathering inputs used in this analysis were obtained through adjusting weathering inputs until predicted surface water outputs for 1984 matched observed values ($R^2 > 0.90$; Fig. 3). The calibrated weathering inputs were used as constant inputs for the simulation period. A paired t test of weathering rates derived in this model application with rates derived through another model application at these sites (Church et al. 1989) showed no significant difference between the two methods.

Changes in watershed base cations

Predicted changes compared to survey data and trends at LTM sites.—The model predicted a general trend of decreases in base cation concentrations in lake waters for the period of 1984–2001. Data from the 2001 survey, however, indicated only a slight decrease or increase in Mg^{2+} concentrations from the 1984 survey (R. A. F. Warby, C. E. Johnson, and C. T. Driscoll, unpublished manuscript). The survey data also indicated larger decreases in Ca^{2+} concentrations than model predictions and the LTM trends (Stoddard et al. 2003). As a result, the predicted concentrations of Ca^{2+} in 2001 were higher than the 2001 survey data and the concentrations of Mg^{2+} were lower (Fig. 3). The predicted median rates of decrease in $\text{Ca}^{2+} + \text{Mg}^{2+}$ during 1990–2000 were $-1.41 \mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$ at NNE and $-1.09 \mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$ at ME, which compared well to the observed LTM trends (median of $-1.48 \mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$; Stoddard et al. 2003).

Predicted changes in base cations in relation to changes in strong acid anions.—The changes in base cations in relation to changes in strong acid anions have implications in recovery of surface water ANC. Based on model calculations, we estimated the ratios of changes in base cations during 1984–2001 to the anions. The calculated ratios were generally above 1, in-

dicating changes in base cations were at similar or faster rates than strong acid anions. In a study by Kirchner (1992), the theoretical stoichiometric ratios of change in base cations in relation to change in acidic anions (F_{SBC}) could be estimated from chemistry of the receiving waters (Kirchner 1992). Using methods described in Kirchner (1992) and lake chemistry data obtained from the 1984 survey, we calculated the theoretical stoichiometric ratios for acid-sensitive sites (i.e., ANC < 50 $\mu\text{eq}/\text{L}$; Table 2). We then calculated expected rates of change in base cations based on the estimated F_{SBC} values and predicted rates of change in strong acid anions. The estimated rates of change in base cations were found to be very similar to predicted rates of changes in base cations ($r = 0.727$). This pattern indicated that at these sites, base cation concentrations decreased at similar or higher rates in response to decreases in strong acid anions, therefore resulting in little or no change in lake ANC. Note that seven out of these 17 acid-sensitive sites showed larger predicted decreases in base cations than the calculated values, indicating depletion of base cation exchangeable pools in soils of at these watersheds might have caused a shift in the stoichiometric ratios.

Changes in drainage loss of base cations in relation to other processes.—Several processes may contribute to decreases in drainage loss of base cations to surface waters, including decreases in base cation deposition, decreases in mineral weathering, decreases in the net release of base cations from the soil exchange complex, and/or increases in net plant uptake. A comparison of simulated changes in fluxes related to these processes during 1984–2001 indicated that decreases in atmospheric deposition generally contributed 20% of the decreases in drainage loss at NNE sites and 40% for ME sites (Fig. 6). The results for NNE agreed with modeling results of Gbondo-Tugbawa and Driscoll (2003), in which they also showed that decreases in deposition contributed 20% of the decreases in base cation drainage loss. The model predictions also showed decreases in mineral weathering inputs, which appeared to be due to decreases in water availability during the simulation period. Changes in the net release of base cations from the soil exchange complex generally accounted for 60–80% of the decreases in drainage loss of base cations in lake waters in NNE and ME. It is difficult to determine if the changes in cation exchange reactions were due to decreases in inputs of mobile anions and/or the depletion of cation exchange pools. However, calculations from section above indicated that for at least seven of the 17 acid-sensitive watersheds, depletion of exchangeable base cation pools was evident. The depletion of exchangeable base cation pools was further supported by changes in simulated soil percent base saturation (%BS). Simulated soil %BS decreases at rates of $-0.02 \pm 0.03\%/yr$ at all the sites, and $-0.05 \pm 0.02\%/yr$ at the acid-sensitive sites of NNE, and $-0.01 \pm 0.04\%/yr$ for all sites and

TABLE 2. Predicted rates of change in the sum of base cation concentrations in relation to changes in strong acid anion concentrations for acid-sensitive watersheds (ANC < 50 $\mu\text{eq/L}$) in NNE and ME.

Site name	D_{CA}^\dagger	D_{CB}^\ddagger	F_{SBC}^\S	$D_{\text{CB}}' \parallel$
Lincoln Pond	-0.64	-0.83	1.06	-0.69
Upper Beech Pond	-1.42	-1.41	1.30	-1.85
Star Lake	-1.58	-1.81	1.16	-1.83
Mendums Pond	-1.10	-1.44	1.07	-1.17
Juggernaut Pond	-2.00	-1.86	0.84	-1.67
Cranberry Pond	-1.69	-2.38	1.27	-2.15
Moore's Pond	-0.97	-1.16	1.16	-1.13
Babbidge Reservoir	-2.00	-2.17	1.02	-2.03
Pemigewasset Lake	-0.63	-1.06	1.19	-0.75
Lt. Greenwood Pond (West)	-1.73	-1.25	1.25	-2.17
Lower Oxbrook Lake	-1.30	-1.69	1.64	-2.13
Duck Lake	-1.83	-2.22	1.27	-2.33
Greenwood Pond	-1.10	-1.16	1.50	-1.65
Long Pond	-0.98	-1.46	1.00	-0.98
Nelson Pond	-2.02	-2.34	0.98	-1.97
Gross Pond	-1.76	-1.76	0.95	-1.68
Kalers Pond	-1.29	-1.54	1.22	-1.57
Mean	-1.41	-1.62	1.17	-1.63
SD	0.46	0.47	0.20	0.51

$\dagger D_{\text{CA}}$ is the predicted rate of change in anions for 1984–2001, calculated as $d\text{NO}_3^- + d\text{Cl}^- + d\text{SO}_4^{2-}$, in $\mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$.

$\ddagger D_{\text{CB}}$ is the predicted rate of change in base cations for 1984–2001, calculated as $d\text{Na}^+ + d\text{Mg}^{2+} + d\text{K}^+ + d\text{Ca}^{2+}$, in $\mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$.

$\S F_{\text{SBC}}$ is the theoretical ratio of changes in base cations relative to acidic anions, calculated as $([\text{Na}^+] + 2[\text{Mg}^{2+}] + [\text{K}^+] + 2[\text{Ca}^{2+}])/([\text{Cl}^-] + [\text{NO}_3^-] + 2[\text{SO}_4^{2-}])$, according to Kirchner (1992). Concentrations are values of 1984 survey data, in $\mu\text{eq/L}$.

$\parallel D_{\text{CB}}'$ is the theoretical rate of change in base cations given the predicted rate of change in anions, calculated as $D_{\text{CA}} \times F_{\text{SBC}}$. Sites with D_{CB} exceeding D_{CB}' (indicated in bold) show depletion of base cations in exchangeable soil pools.

$-0.05 \pm 0.02\%/yr$ for acid-sensitive sites in at ME. Effects of biotic processes were generally negligible in the mass balances of base cations at NNE sites (i.e., net mineralization, net plant uptake). However, model simulation indicated a net release of base cations from biotic processes at ME sites. This release was probably a result of colder temperature at ME than NNE, which might result in less plant production and demand for base cations.

SIMULATIONS IN SURFACE WATER ANC AND RESPONSE TO FUTURE CONTROL SCENARIOS

Simulations in surface water ANC

Predicted lake ANC in 1984 and 2001 compared well with survey data (Fig. 3). The model predicted minor increases in surface water ANC at NNE sites of $0.46 \pm 0.78 \mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$ and a continuous decrease in ANC at ME sites of $0.48 \pm 1.23 \mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$. These rates of change compared well to trends observed in the LTM sites in NNE and ME, with a median rate of $0.11 \mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$ (Stoddard et al. 2003). For sites in NNE and ME, declines in SO_4^{2-} have been largely offset by decreases in base cation concentrations, thus resulting in little or no change in ANC, as indicated by model simulations and the calculated F_{SBC} values shown above.

Historical acidification

Model hindcasts of lake and soil chemistry prior to the onset of marked increases in SO_2 and NO_x emissions (circa 1850) help show the impacts of acidic deposition as well as establish a baseline of reference conditions upon which to evaluate the extent of recovery. In our analysis, the model hindcasts were conducted under the same conditions as simulation 4 (i.e., with all the corrections in S simulations). Our analysis showed that prior to increases in acidic deposition, lake concentrations of SO_4^{2-} and NO_3^- were low across the subregions. Simulated background SO_4^{2-} concentrations of these two subregions were generally less than $20 \mu\text{eq/L}$, with median values of $8\text{--}9 \mu\text{eq/L}$. Simulated concentrations of NO_3^- prior to increases in acidic deposition were generally less than $1 \mu\text{eq/L}$ in 1850.

Calculated background pH and ANC in 1850 have median values of 6.7 and $116 \mu\text{eq/L}$ for NNE and 6.7 and $109 \mu\text{eq/L}$ for ME, respectively. For current acid-sensitive watersheds (i.e., $\text{ANC} < 50 \mu\text{eq/L}$), the model estimated median background pH and ANC as 6.3 and $56.3 \mu\text{eq/L}$, respectively. For acid-sensitive watersheds, predicted background concentration of inorganic monomeric Al was $0.09 \mu\text{mol/L}$.

Response to future scenarios

Future scenarios.—Current emission reduction proposals call for 50–75% reductions in emissions of SO_2

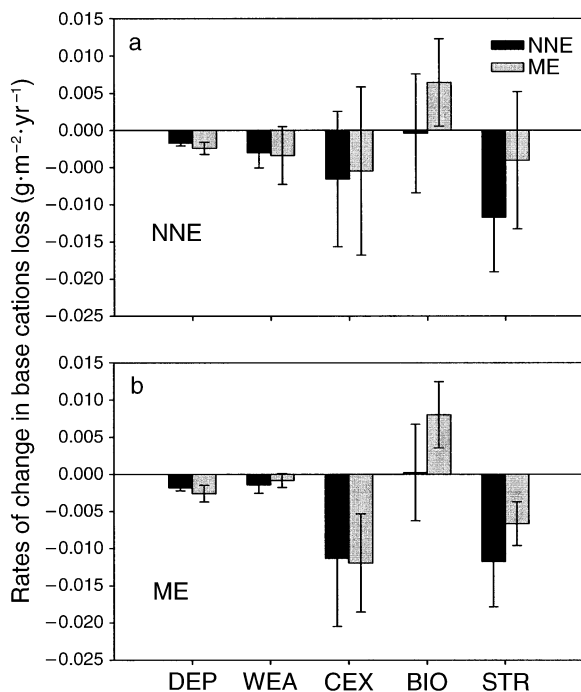


FIG. 6. Estimated rates of change in base cation deposition (DEP) in relation to rates of change in mineral weathering (WEA), net cation exchange (CEX), net effect of biotic processes (BIO), and stream loss of base cations (STR) for (a) all sites and (b) acid-sensitive sites (ANC < 50 $\mu\text{eq/L}$) in northern New England (NNE) and Maine (ME) for the period 1984–2001. Values are mean \pm SD.

from electric utilities beyond full implementation of Title IV of the 1990 CAAA and 70–75% reduction from 1997 levels in NO_x emissions to be implemented by various dates (Driscoll et al. 2001). To evaluate effects of possible future emission controls, we considered three future control scenarios of (1) the base case (implementation of the 1990 CAAA); (2) the moderate control; and (3) the aggressive control (Table 3). These reductions in emissions were assumed to be implemented linearly during 2010–2020. Similar to the model hindcasts, the simulations for future scenarios were conducted under the same conditions of simulation 4 of the S analysis (see Table 1). Simulations for future scenarios were conducted also under the assumption

that deposition of base cations and NH_4^+ remains as the average values of the 1990s. We also assumed constant climate for the future scenarios.

Extent of recovery.—Predicted lake SO_4^{2-} concentrations decrease under all three future scenarios for the period 2010–2050 (Fig. 7). The rates of decrease in SO_4^{2-} are generally uniform under each scenario and additional controls accelerate the declines of SO_4^{2-} . The rates of decline are slightly greater at NNE sites than ME sites. This pattern could be due to the fact that the same amount reduction of SO_2 emissions would likely result in larger decreases in S deposition at the NNE sites that are closer to the source area than ME sites. Predicted SO_4^{2-} concentrations for 2050 are also relatively uniform under each scenario and are generally less than 60 $\mu\text{eq/L}$ (Fig. 8). Predicted SO_4^{2-} concentrations under the aggressive control scenario are generally less than 30 $\mu\text{eq/L}$ for both NNE and ME sites, but still considerably greater than our estimated background values.

The relatively rapid declines in SO_4^{2-} will result in increases in ANC at most of the lakes in both NNE and ME. Under the base case scenario, predicted median rates of increase in ANC are about 0.03 $\mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$ at NNE and 0.02 $\mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$ at ME. Additional control greatly enhances the recovery of ANC to 0.30 $\mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$ at NNE and 0.22 $\mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$ at ME under the aggressive control scenario. These results are similar to the predictions in the Adirondacks, although predicted median rates of ANC increase are slightly higher in the Adirondacks (0.4 $\mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$ under the aggressive control scenario; Chen and Driscoll 2005). For soils, the decreases in SO_4^{2-} will generally result in increases of soil %BS (Fig. 7). However, the rates of increase in soil %BS are relatively small, suggesting ongoing depletion of available nutrient cation pools.

Status in 2050.—Critical chemical thresholds that appear to coincide with the onset of deleterious effects to biotic resources include (1) the molar Ca/Al ratio of soil water < 1.0 and soil percent base saturation < 20%, which indicate that forest vegetation is at risk with respect to soil acidification from acidic deposition (Cronan and Schofield 1990, Cronan and Grigal 1995); and 2) surface water pH < 6.0, ANC < 50 $\mu\text{eq/L}$ and

TABLE 3. Summary of the three future scenarios used in model simulations.

Model	Summary
1990 CAAA (base case)	SO_2 emissions from utility capped at 8.95 million short tons (8.12×10^6 Mg). Non-utility SO_2 emissions capped at 5.6 million short tons (5.08×10^6 Mg). Overall ~40% reduction in SO_2 emissions achieved by 2010 (through Phase I and Phase II), relative to 1990 emissions. Overall ~5% reduction in NO_x emissions by 2010.
Moderate control	Reduce the utility SO_2 emissions to 6 million short tons (5.44×10^6 Mg); ~20% reduction in annual NO_x emissions and ~55% reduction in SO_2 emissions by 2010, relative to 1990.
Aggressive control	Reduce the utility SO_2 emissions to 1 million short tons (0.91×10^6 Mg); ~30% reduction in annual NO_x emissions and ~75% reduction in SO_2 emissions by 2010, relative to 1990.

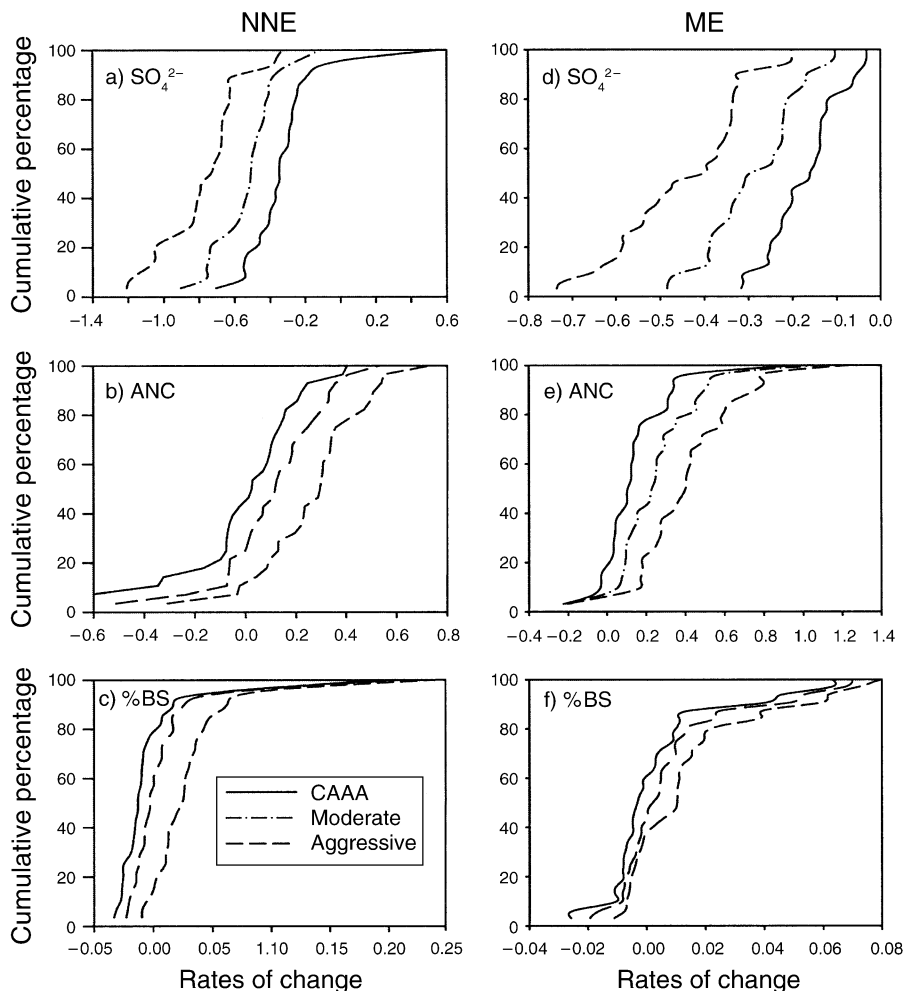


FIG. 7. Distribution of predicted rates of changes in surface water SO_4^{2-} concentrations ($\mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$), ANC ($\mu\text{eq}\cdot\text{L}^{-1}\cdot\text{yr}^{-1}$), and soil %BS (percent base saturation, %/yr) during 2001–2050 under the three scenarios of base case (1990 CAAA), moderate control, and aggressive control at (a–c) northern New England (NNE) sites and (d–f) Maine (ME) sites.

inorganic monomeric Al concentrations $>2 \mu\text{mol/L}$, which indicate that aquatic biota are at risk from surface water acidification due to acidic deposition (McAvoy and Bulger 1995).

Predicted soil solution Ca/Al ratios in 2050 are generally higher than the critical values of 1 for sites at both subregions. Soil %BS at 80% of the NNE sites and 60% of the ME sites are generally less than critical value of 20%. Median background %BS is 26.8% at ME and 22.2% at NNE.

Since most of the DDRP sites in these two subregions had ANC values greater than $50 \mu\text{eq/L}$ in 1984, predicted surface water pH and ANC in 2050 are generally above the critical values, and for inorganic monomeric Al concentrations below the critical values. However, about 20% of the sites will have pH less than 6, and ANC of around 25% of the watersheds will remain under $50 \mu\text{eq/L}$ in 2050.

CONCLUSIONS

We summarize the main findings from this study:

1) For both northern New England and Maine, elevation appeared to be an important factor influencing inputs of atmospheric S deposition to watersheds. Including elevation as a component in estimating S deposition inputs significantly improved model predictions in lake SO_4^{2-} concentrations.

2) Changes in lake SO_4^{2-} concentrations were related to changes in atmospheric deposition, although desorption of soil SO_4^{2-} delayed the response of lake water to decreases in atmospheric S deposition.

3) The declines in $\text{SO}_4^{2-} + \text{NO}_3^-$ concentrations were coupled with stoichiometric decreases in base cation concentrations, as indicated by the model predictions. As a result, changes in surface water ANC in response to changes in atmospheric deposition were minor.

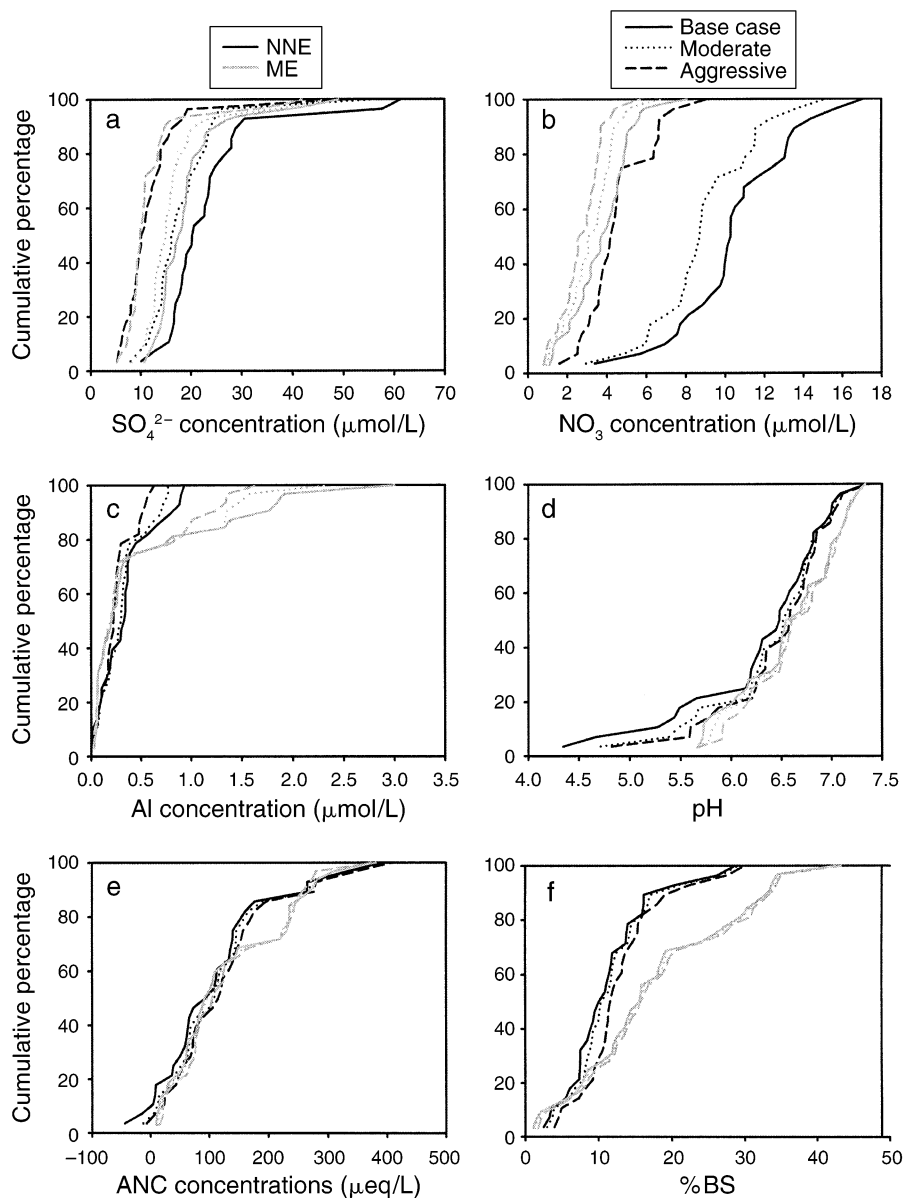


FIG. 8. Predicted surface water (a) SO_4^{2-} concentrations, (b) NO_3^- concentrations, (c) inorganic Al concentration, (d) pH, (e) ANC, and (f) soil %BS (percent base saturation) in 2050 under three scenarios for northern New England (NNE) and Maine (ME) sites.

4) Decreases in base cation deposition contributed an average of 20% and 40% to decreases in drainage loss of base cations for sites in NNE and ME, respectively. The decreases in lake base cation concentrations were largely due to declines in the net release of basic cations from the soil exchange complex.

5) Future reductions in S deposition will contribute to the recovery in soil and surface waters. Although marked improvement is expected, chemical and biological recovery to the background conditions will not be expected at all the sites in 2050.

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