Ecological Applications, 3(3), 1993, pp. 459–472

A SPATIAL MODEL OF ATMOSPHERIC DEPOSITION FOR THE NORTHEASTERN U.S.[1]

SCOTT V. OLLINGER AND JOHN D. ABER
Complex Systems Research Center, Institute for the Study of Earth, Oceans and Space,
University of New Hampshire, Durham, New Hampshire 03824 USA

GARY M. LOVETT
Institute of Ecosystem Studies, The New York Botanical Garden, Mary Flagler Cary Arboretum,
Millbrook, New York 12545 USA

SARAH E. MILLHAM[2]
Complex Systems Research Center, Institute for the Study of Earth, Oceans and Space,
University of New Hampshire, Durham, New Hampshire 03824 USA

RICHARD G. LATHROP
Department of Environmental Resources, Cook College—Rutgers University,
New Brunswick, New Jersey 08903 USA

JENNIFER M. ELLIS
Complex Systems Research Center, Institute for the Study of Earth, Oceans and Space,
University of New Hampshire, Durham, New Hampshire 03824 USA

Abstract. Spatial patterns of atmospheric deposition across the northeastern United States were evaluated and summarized in a simple model as a function of elevation and geographic position within the region. For wet deposition, 3–11 yr of annual concentration data for the major ions in precipitation were obtained from the National Atmospheric Deposition Program/National Trends Network (NADP/NTN) for 26 sites within the region. Concentration trends were evaluated by regression of annual mean concentrations against latitude and longitude. For nitrate, sulfate, and ammonium concentrations, a more than twofold linear decrease occurs from western New York and Pennsylvania to eastern Maine. These trends were combined with regional and elevational trends of precipitation amount, obtained from 30-yr records of annual precipitation at >300 weather stations, to provide long-term patterns of wet deposition.

Regional trends of dry deposition of N and S compounds were determined using 2–3 yr of particle and gas concentration data collected by the National Dry Deposition Network (NDDN) and several other sources, in combination with estimates of deposition velocities. Contrary to wet deposition trends, the dominant air concentration trends were steep decreases from south to north, creating regional decreases in total deposition (wet + dry) from the southwest to the northeast.

This contrast between wet and dry deposition trends suggests that within the northeast the two deposition forms are received in different proportions from different source areas, wet deposited materials primarily from areas to the west and dry deposited materials primarily from urban areas along the southern edge of the region.

The equations generated describing spatial patterns of wet and dry deposition within the region were entered into a geographic information system (GIS) containing a digital elevation model (DEM) in order to develop spatially explicit predictions of atmospheric deposition for the region.

Key words: cations; dry deposition; elevation; geographic information system; nitrogen; precipitation; sulfur; wet deposition.

INTRODUCTION

Anthropogenic increases in atmospheric deposition can have profound and possibly adverse effects on terrestrial and aquatic ecosystems (Cronan and Schofield 1980, Reuss and Johnson 1986, Aber et al. 1989, Schulze 1989). Of particular concern are increases in the deposition of SO_4^{2-} and NO_3^-, which can affect natural systems through their activity both as mobile anions and essential nutrients.

Anion effects include increased leaching of nutrient cations (Ca^{2+}, Mg^{2+} Na^+, K^+), especially in soils with low SO_4^{2-} adsorption capacity, and leaching of aluminum species and hydrogen ions in soils with low

---

[1] Manuscript received 6 April 1992; revised 1 November 1992; accepted 5 November 1992.

[2] Present address: Department of Geography, University of Maryland, College Park, Maryland 20742 USA.

base saturation (Cronan and Schofield 1980, Reuss and Johnson 1986). Nutrient effects primarily concern inputs of nitrogen. Although small increases in N deposition can produce moderate increases in forest productivity, excessive N loading may have several adverse effects on both forest health and water quality (Friedland et al. 1984, Nihlgard 1985, van Breeman and van Dijk 1988, Aber et al. 1989, Schulze 1989).

Atmospheric deposition occurs via several mechanisms: wet deposition (rain and snow), dry deposition (direct deposition of particles and gases), and cloud water deposition (direct deposition of cloud and fog droplets). Of these, wet deposition is by far the most extensively measured, although dry deposition may be as important as wet deposition for sulphur and nitrogen, and cloud water deposition can dominate in mountaintop sites (Lovett and Kinsman 1990).

Spatial variation in atmospheric deposition occurs over several different scales. At local scales, topography and vegetation canopy characteristics can cause significant variation in deposition rates, especially in mountainous landscapes (Weathers et al. 1992). Topography affects wet deposition primarily by orographic uplifting of air masses, which causes precipitation to increase with elevation (Barrie 1981, Lovett and Kinsman 1990). At high elevations, wet deposition can be further increased by the scavenging of cloud droplets by falling rain or snow (Fowler et al. 1988). Additional increases in total deposition at high elevations result from direct cloud water inputs (Lovett et al. 1982) and dry deposition, both of which are thought to increase with elevation (Lovett and Kinsman 1990).

At larger scales, patterns of both wet and dry deposition result from the locations of source areas relative to the dominant patterns of atmospheric circulation. For wet deposition, spatial patterns have been assessed using data from several widely dispersed wet deposition monitoring networks (Wilson and Mohnen 1982, Munger and Eisenreich 1983, Summers et al. 1986, Wisniewski and Kinsman 1988, Zemba et al. 1988). Continental-scale contour maps generated from these studies have indicated that in the U.S., maximum wet deposition of SO_4^{2-}, NO_3^-, and H^+ occurs around the lower Great Lakes region. Deposition rates then tend to decrease slowly over distances of hundreds to thousands of kilometres as air masses move away from the source zone and become depleted of suspended materials.

Atmospheric concentrations of S and N oxides show a similar pattern, although there are fewer measurement sites and the data record covers a shorter time period (Edgerton and Lavery 1991). Dry deposition patterns have not been measured directly, but model calculations indicate that the deposition patterns should be similar to the atmospheric concentration patterns over a regional scale (Edgerton and Lavery 1991).

Although existing deposition contour maps show general trends at a regional scale, predicting deposition to a particular site is difficult since only one to a few isolines generally pass through a given region. These maps also offer no way to incorporate the local effects of topography, which can be significant in mountainous areas.

The objectives of this study were (1) to develop equations summarizing existing data on precipitation quantity and chemistry across the northeastern U.S. as a function of elevation and geographic location within the region, (2) to develop similar equations describing geographic patterns of particle and gas concentrations, and to use these data to estimate regional patterns of dry deposition, and (3) to use these equations within a geographic information system (GIS) to develop spatially explicit predictions of total deposition for the region.

METHODS

Wet deposition

Previous studies of wet deposition patterns have generally used precipitation amounts from only the sites where deposition samples were collected to determine patterns of deposition amount. This greatly limits both the number of data points from which regional patterns can be generated, and the length of the time series of data available. In this study, we derive spatial patterns of ion concentration in wet deposition, and then combine them with long-term precipitation data from 310 weather stations in order to generate maps with higher spatial resolution of wet deposition.

Ion concentration.—Data representing volume-weighted mean annual concentrations of eight major ions (Ca^{2+}, Mg^{2+}, K^+, Na^+, NH_4^+, NO_3^-, Cl^-, and SO_4^{2-}) in wet deposition for 30 sites across the northeastern U.S. (from Pennsylvania to Maine) were obtained from the National Atmospheric Deposition Program/National Trends Network (NADP/NTN 1991). Sites having <3 yr of data were omitted. Completeness criteria established by NADP/NTN were used to determine which data were suitable for use in computing long-term mean concentrations: (1) there must have been valid precipitation chemistry data for at least 75% of both the summary period and the precipitation reported, and (2) the volume of each sample collected must have been at least 75% of that expected based on rain gauge precipitation measurements. Data that did not meet these criteria were rejected. The remaining data set included 26 sites (Fig. 1), each containing from 3 to 11 yr of data (mean = 6.7 yr). Annual means were then used to compute long-term, volume-weighted mean concentrations for each location.

All of the NADP/NTN collection sites are located in rural areas to avoid inputs from local pollution sources. Although this allows clearer identification of large-scale trends, the effects of local sources, which can be substantial in urban or industrial areas, cannot be accounted for.

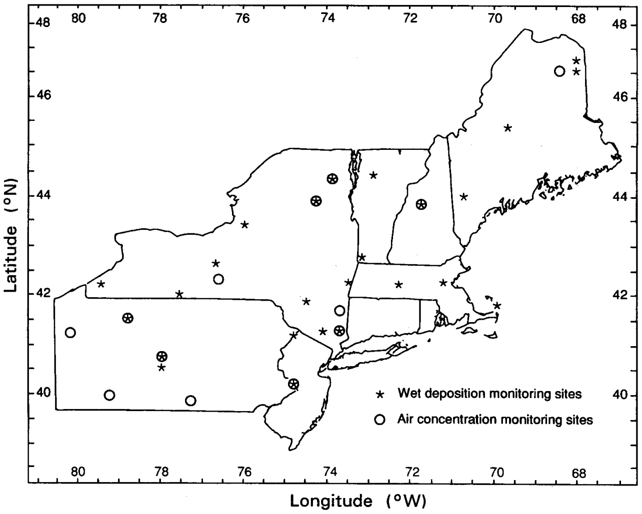


FIG. 1. Locations of the 26 precipitation chemistry and 13 air concentration monitoring stations used for evaluating regional trends in the concentrations of wet- and dry-deposited species.

Regional trends in ion concentration were evaluated by linear regression analyses of concentration against latitude and longitude (in decimal degrees) as well as multiple regressions against both. Regressions were also performed with elevation included, although the elevation range of the NADP/NTN data set is limited (18–634 m). In addition to spatial variables, mean annual precipitation amounts were included in the regressions to determine whether a dilution–enrichment effect could contribute to, or mask, spatial trends. Since a slight decrease in wet deposition of sulfur and several other ions has occurred over the past decade (Hedin et al. 1987, Driscoll and Van Dreason, *in press*), we also examined relationships between the time periods of data collection at the NADP/NTN sites and their locations within the region in order to determine whether temporal trends could influence the detection of spatial trends.

Precipitation amount. — Data representing long-term (30-yr) mean annual precipitation for 307 locations across New England and New York state were obtained from the National Oceanic and Atmospheric Administration (NOAA 1982). The sites were evenly distributed across the region and ranged in elevation from 2 to 618 m, with the exception of Mount Washington, New Hampshire at 1909 m. Since higher elevation sites were poorly represented in the data set, mean annual precipitation data from several other high elevation sites were added. These include 808 m on Slide Mt., New York (from 7 yr of data), 1205 m on Mt. Mansfield, Vermont (from 11 yr of data; the above two sites operated by NOAA), and 900 m at the Hubbard Brook Experimental Forest in New Hampshire (from 30 yr of data; operated by the USDA Forest Service, Federer et al. 1990).

It was anticipated that most of the spatial variation in precipitation would occur at two levels, (1) a regional scale that could be expressed in terms of latitude and/or longitude, and (2) a local scale, being a function of elevation. Analysis of spatial patterns was thus performed by regression analysis of precipitation against latitude, longitude, and elevation.

Dry deposition

Since the direct measurement of dry deposition is difficult, few reliable dry deposition data are available. As an alternative, we use an inferential method whereby atmospheric concentrations of dry-deposited species are combined with estimates of deposition velocities in order to estimate dry deposition amounts.

Atmospheric concentrations. — Data on atmospheric concentrations of the gases SO_2 and HNO_3 and the particulate species NO_3^-, SO_4^{2-}, and NH_4^+ in New England, New York, Pennsylvania, and New Jersey were obtained from several sources. Annual mean data from 11 sites for 1989 and 1990 were obtained from the E.P.A.-sponsored National Dry Deposition Network (NDDN, Edgerton et al. 1991, Edgerton and Lavery 1991). Measurements were made using a filter-pack composed of teflon-, nylon-, and sodium carbonate-coated filters in series in a flow-controlled sampling line. Two other sites that obtained data using the same methods were included: the Huntington Forest site in the eastern Adirondacks (1986–1988 mean concentrations reported by Johnson and Lindberg 1992) and the Institute of Ecosystem Studies site in southeastern New

York (1988–1990 mean concentrations, IES 1988, 1989, 1990).

As was the case for precipitation chemistry stations, all air concentration sites are located in rural areas to avoid the effects of local sources. Again, this might allow easier identification of large-scale trends, but important local effects may remain undetected. A particularly important example of this problem is nitrogen dioxide (NO_2) which may cause large increases in N deposition near urban areas (Hanson et al. 1989), but is not measured at the air concentration monitoring sites used in this study. Likewise, NH_3 and NO may be locally important near sources, but concentration data are not available.

As was done for precipitation chemistry, we evaluated regional trends by linear regression analyses of mean annual concentrations against latitude and longitude. This data set is inferior to the precipitation chemistry data set because only 13 sites are available and the data record at each site is only 2–3 yr. The data are also insufficient for resolving any elevation effects. In addition, the chemical species measured permit the analysis of only N and S deposition. Nonetheless, there is sufficient information available to begin to discern regional patterns of atmospheric concentrations and contrast them to precipitation chemistry and wet deposition.

Deposition velocities.—To calculate dry deposition fluxes from atmospheric concentrations, dry deposition velocities must be specified. Deposition velocities are difficult to measure and are variable in magnitude, depending on the nature of the depositing substance, the vegetation, and meteorological factors. Because deposition velocities have not been measured at most of the sites from which these atmospheric concentration data were obtained, we cannot directly evaluate patterns of dry deposition. However, considering the study area as a whole, the factors most likely to produce changes in dry deposition across the region are (1) variation in the vegetation cover, (2) variation in meteorological conditions, and (3) variation in atmospheric concentrations. If we restrict our predictions to the same vegetation type (e.g., mature deciduous forest) and assume that spatially and temporally averaged meteorological conditions (e.g., wind speed) do not vary substantially across the region, then the patterns in atmospheric concentrations will reflect patterns of dry deposition. This assumption is supported by Edgerton and Lavery (1991) who calculated that changes in deposition of SO_2 across the eastern United States are influenced to a much greater extent by concentration differences than by differences in deposition velocity.

To estimate potential magnitudes of dry deposition, we chose representative deposition velocities for the different chemical species and applied them to the measured concentrations across the study region. Following Lindberg et al. (1986), we chose deposition velocities characteristic of a deciduous forest (averaging growing and dormant season values for an annual mean) of 1.3 cm/s for HNO_3 and 0.13 cm/s for particulate $SO_4{}^{2-}$, $NO_3{}^-$, and $NH_4{}^+$. For SO_2, we chose a deposition velocity of 0.22 cm/s by taking the average of values calculated by Edgerton and Lavery (1991) for several sites across the region. These are our best estimates of the dry deposition velocities, but they may be inaccurate by as much as a factor of 2. These deposition velocities were multiplied by the appropriate concentrations to determine approximate dry deposition fluxes.

Geographic information system

A digital elevation model (DEM) was obtained from the U.S. Geologic Survey (USGS 1987) and loaded into the Geographic Resources Analysis Support System (GRASS 4.0), a raster-based GIS software program (Westerfelt et al. 1987). The DEM covers eastern New York and New England with a geographic resolution of 30 arc-seconds. Wet deposition fields were generated by processing each pixel according to the appropriate equations for ion concentration and precipitation amount discussed below. Dry deposition fields were then added using similar equations for atmospheric concentrations along with the deposition velocity estimates discussed above.

RESULTS

Ion concentrations in precipitation

Results of regressions describing regional variation in ion concentration are presented in Table 1. Several ions show significant increases in concentration from east to west across the region (Fig. 2). For $NO_3{}^-$, $SO_4{}^{2-}$, H^+, Ca^{2+}, and K^+, the regional trends observed are best described as functions of longitude alone. Including latitude in the analyses did not significantly improve any of the relationships. Latitude is, however, a statistically significant predictor for $NH_4{}^+$, resulting in an increase in $NH_4{}^+$ concentration from the southeast to the northwest within the region. No correlation was found between the periods of data collection at the NADP/NTN sites and their locations within the region, indicating that the slight decrease in wet $SO_4{}^{2-}$ deposition that has occurred over the period of data collection (Hedin et al. 1987, Driscoll and Van Dreason, *in press*) has not contributed to the $SO_4{}^{2-}$ concentration gradient.

Concentrations of Na^+, Cl^-, Mg^{2+}, and K^+ were greatly elevated at two coastal sites (N. Atlantic Coastal Lab, Barnstable, Massachusetts and Acadia National Park, Maine) presumably due to inputs from sea spray. These ions were ≈5 and 2.5 times more concentrated at the two respective coastal sites than at sites only slightly further inland. Data from these coastal sites were, thus, omitted. For Na^+ and Cl^-, the trends from the remaining sites are best described as logarithmic decreases in concentration with distance from the ocean

TABLE 1. Regression equations describing regional patterns of ion concentrations in precipitation (in mg/L, $n = 26$). All coefficients are significant at $P < .05$ except the longitude Mg^{2+} coefficient for which $P < .10$. Standard errors are in parentheses.

	Coefficients				
Ion	Constant	Longitude	Latitude	Adjusted R^2	P
SO_4^{2-}	−9.5871	0.1612		0.83	<.0001
	(1.06)	(0.01)			
NO_3^-	−7.0148	0.1152		0.81	<.0001
	(0.83)	(0.01)			
NH_4^+	−2.4359	0.0259	0.0172	0.78	<.0001
	(0.43)	(0.003)	(0.006)		
H^+	−0.2205	0.0036		0.84	<.0001
	(0.02)	(0.0003)			
Ca^{2+}	−0.3804	0.0066		0.41	<.001
	(0.11)	(0.002)			
K^+*	−0.0439	0.0009		0.21	.01
	(0.02)	(0.0006)			
Mg^{2+}*	0.3488	−0.0017	−0.0046	0.24	.02
	(0.05)	(0.001)	(0.002)		
ln Na^+*	26.4011	−0.1932	−0.3372	0.58	<.0001
	(4.91)	(0.04)	(0.06)		
ln Cl^-*	24.8873	−0.1604	−0.3427	0.56	<.0001
	(4.90)	(0.04)	(0.06)		

* Regressions performed with two coastal sites omitted.

that can be approximated by regressing log-transformed concentration values on latitude and longitude. Predictions from these equations do not account for the local increases expected at sites immediately adjacent to the seacoast, but do fit the existing data for inland sites better than regressions that include the coastal sites. Magnesium concentrations also decrease with distance from the ocean, but the trend is weaker and is best fit as a linear function of latitude and longitude.

Given the limited elevational distribution of the NADP/NTN collection sites, the precipitation chemistry data were thought to be inadequate for meaningful analysis of elevation trends in ion concentration. Including elevation in the regressions (along with the variables listed in Table 1), however, produced significant effects for most ions. The strengths of the effects varied greatly from ion to ion, but all showed a decrease in concentration with increasing elevation. The elevation coefficients (in milligrams per litre per metre) were as follows: $SO_4^{2-} = -0.00089$; $NO_3^- = -0.00044$; $H^+ = -0.000019$; $NH_4^+ = -0.00016$; $Ca^{2+} = -0.000058$; $K^+ = -0.000018$; $Mg^{2+} = -0.00035$; $Na^+ = -0.00018$; and $Cl^- = -0.00039$ (Na^+ and Cl^- coefficients apply to untransformed values). All of the above coefficients are significant at the .05 level except NO_3^- ($P = .07$) and Ca^{2+} ($P = .08$), although most did not substantially improve the fit of the equations (most adjusted R^2 values increased by no more than 0.05).

For NO_3^-, SO_4^{2-}, and H^+, these elevation effects are relatively small if applied to the narrow elevation range that comprises most of the region. Projecting these effects to higher elevations, however, could produce substantial, but questionable results. For example, at longitude = 72°, concentrations of NO_3^-, SO_4^{2-}, and H^+ would decrease by 33%, 43%, and 38% respectively from 200 to 1200 m in elevation, causing sizeable reductions in wet deposition estimates. Other ions would show greater, and perhaps even unrealistic, decreases. Over the same elevation range (200–1200 m), Ca^{2+} concentrations would decrease by 59%, Mg^{2+} by 94%, K^+ by 98%, Na^+ and Cl^- by 78% and 82% respectively, and NH_4^+ by 188%.

One factor that could cause these apparent elevational trends is the increase in precipitation amount experienced at higher elevations, possibly causing a dilution of ion concentrations. Within the data collected by NADP/NTN, however, there were no significant correlations between mean annual concentration and mean annual precipitation amount for any of the ions. Additionally, other studies conducted in the eastern U.S. that were more suitably designed for observing elevational trends found no significant changes in the concentrations of NO_3^-, SO_4^{2-}, and H^+ with elevation (Tennessee Valley Authority 1983, Scherbatskoy and Bliss 1984, Lindberg et al. 1988). It should also be noted that within the NADP/NTN data set, elevation increases to the north and west, a factor which further confounds attempts to identify elevation trends. It was thus decided that elevational trends in ion concentration should not be included in the model unless future work more confidently demonstrates the existence of such trends.

Precipitation amount

Regression of precipitation amount against latitude and longitude revealed a linear decrease in precipita-

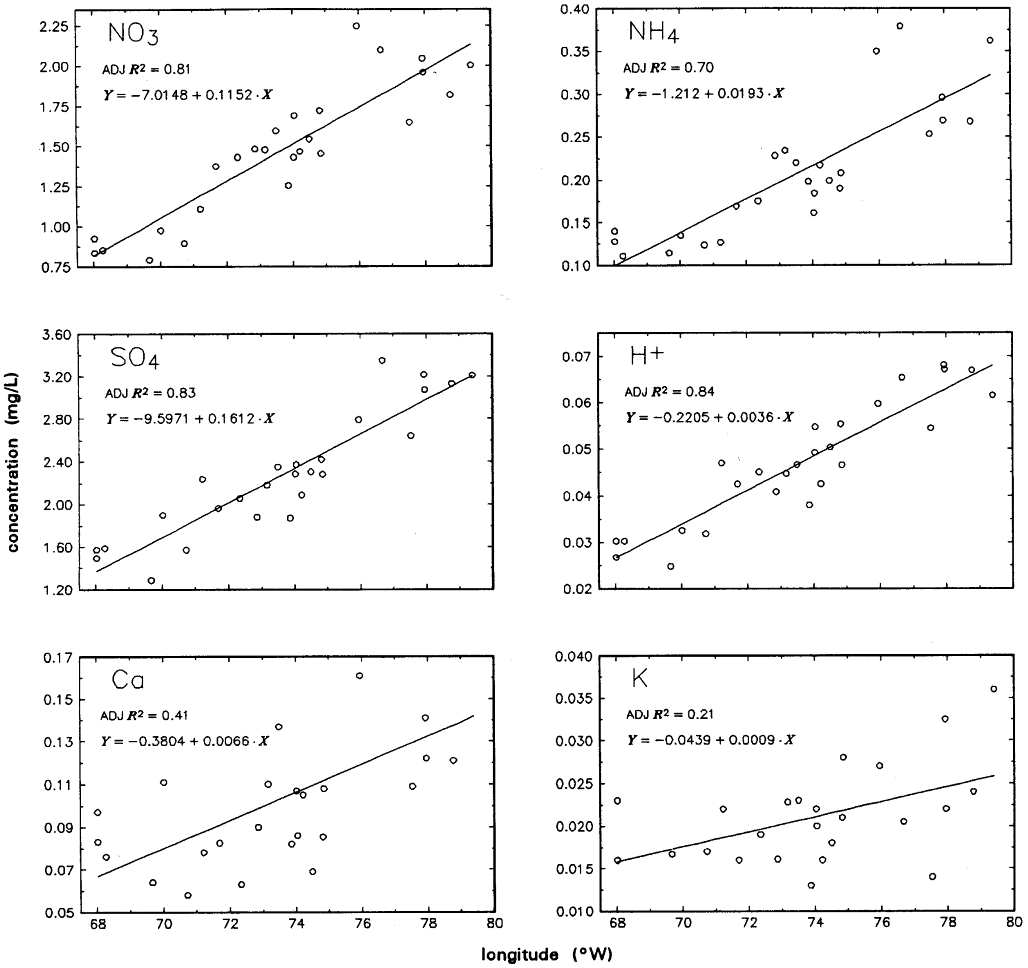


FIG. 2. Regression plots of mean annual concentration against longitude for ions in precipitation. Note that a slight latitude trend is also present in the NH_4^+ data (Table 1).

tion from the southeast to the northwest (approximating a trend of decreasing precipitation with increasing distance from the ocean). A plot of residuals from this regression against elevation (which approximates the elevation trend in precipitation) shows a discontinuity at ≈400 m, above which there is a strong, linear increase, and below which there is only a mild elevation effect (Fig. 3). To fit this pattern with a regression, slope and intercept dummy variables were created as described by Hamilton (1990). The intercept variable (IV) was given a value of 0 for all sites below 400 m and 1 for all sites above 400 m. The slope variable (SV) was created by multiplying IV by elevation. Precipitation was then regressed against latitude, longitude, elevation, and the two new variables. The adjusted R^2 value for the regression was 0.65 (root MSE = 9.904 cm), and all coefficients were significant to the .05 level, indicating that the relationship between precipitation (ppt, in centimetres) and elevation (elev, in metres) differs significantly above and below 400 m.

For all locations below 400 m, using IV = 0 reduces the regression equation to:

$$\text{ppt} = 670.20 - (7.02 \cdot \text{latitude}) - (3.62 \cdot \text{longitude}) + (0.013 \cdot \text{elev}), \quad (1)$$

and above 400 m, using IV = 1 gives:

$$\text{ppt} = 643.43 - (7.02 \cdot \text{latitude}) - (3.62 \cdot \text{longitude}) + (0.090 \cdot \text{elev}). \quad (2)$$

The elevation coefficients indicate a 13 cm/1000 m increase in precipitation with elevation at low elevations and a 90 cm/1000 m increase above 400 m.

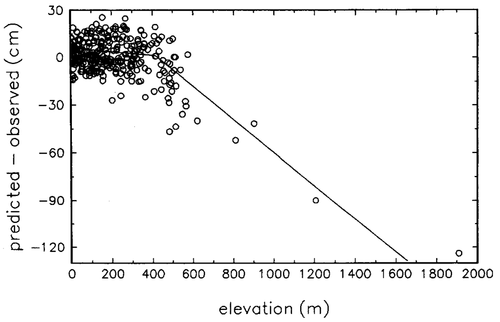


FIG. 3. Variation of annual precipitation with elevation, as shown by plotting residuals vs. elevation from a regression of mean annual precipitation against latitude and longitude (residuals = predicted − observed values, $n = 310$). The line represents the elevation effect described by a dummy variable regression indicating a significant discontinuity at $\approx$400 m.

Total wet deposition

Estimates of mean annual wet deposition to a site are determined by multiplying the predicted value of ion concentration (Table 1) by the predicted annual precipitation amount (Eqs. 1 and 2) per unit land area. Fig. 4 shows plots of predicted vs. observed wet deposition of N and S for the 26 NADP/NTN stations used. Since data from these sites were used to produce the concentration equations, this is not a true validation exercise. These plots do, however, include error introduced by the precipitation equations as well as residuals from the concentration regressions.

In general, predictions agree very well with observed values. Two exceptions were sites at Bennett Bridge, New York and Jasper, New York. Measured values at Bennett Bridge were $\approx$1.5 and 1.8 times higher than predictions for N and S, due to differences in both precipitation and ion concentrations. Mean annual precipitation for the site was 131 cm, considerably higher than the predicted value of 92 cm. The large amount of precipitation typically received at the site is presumably due to its location on Lake Ontario, a factor which is not accounted for in model predictions. Measured concentrations of N and S at Bennett Bridge were also high. Residual values from the regressions shown in Fig. 2 were greater at Bennett Brook than any other site, possibly indicating an influence of local emission sources.

In contrast, predicted values of N and S for Jasper, New York (located in southcentral New York state) were nearly two times higher than measured values, mostly due to the low average precipitation amount measured at the site (72 cm/yr) compared to the predicted value of 105 cm/yr. This may also reflect local-scale variation in precipitation that cannot be accounted for in the region-wide regressions performed. The exact location of the precipitation collector at Jasper might reveal a microtopographic effect (e.g., rain shadowing) responsible for the unusually low amounts of precipitation recorded there.

Predicted annual wet deposition of NO_3^- at high elevations (>1200 m) in the Adirondack Mountains of New York can be as much as 30 kg·ha^{-1}·yr^{-1}, twice that expected at adjacent low elevation sites (300 m) and three times that expected at low elevations in eastern Maine. With the additional input of NH_4^+, total wet nitrogen (NO_3^--N + NH_4^+-N) deposition at low elevations is predicted to range from $\approx$6.6 kg·ha^{-1}·yr^{-1} in western New York to 3.0 kg·ha^{-1}·yr^{-1} in Maine. Total wet sulfur deposition (as elemental S) decreases from 9.4 to 4.3 kg·ha^{-1}·yr^{-1} over the same range.

Relatively few high elevation data are available for comparison with predictions. Miller et al. (*in press*) measured an average wet deposition flux of 19.22 kg·ha^{-1}·yr^{-1} of NO_3^-, 25.4 kg·ha^{-1}·yr^{-1} of SO_4^{2-}, and 3.41 kg·ha^{-1}·yr^{-1} of NH_4^+ over a 4-yr period at a 1050 m site on Whiteface Mt., New York. Model predictions for the site are 22.8 kg·ha^{-1}·yr^{-1} for NO_3^-, 35.0 kg·ha^{-1}·yr^{-1} for SO_4^{2-}, and 3.65 kg·ha^{-1}·yr^{-1} for NH_4^+. These predictions are based on precipitation of 153 cm/yr (calculated from Eq. 2), which is somewhat higher than the average of 132 cm/yr measured over 4 yr at the Whiteface site. Concentrations measured at Whiteface were 1.45, 1.93, and 0.258 mg/L

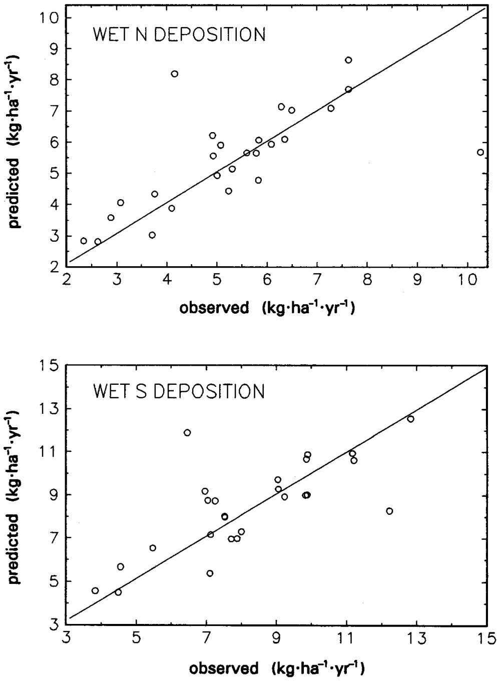


FIG. 4. Plots of predicted vs. observed wet deposition of nitrogen (from NO_3^- + NH_4^+) and sulfur at the 26 NADP/NTN collection stations used in this study.

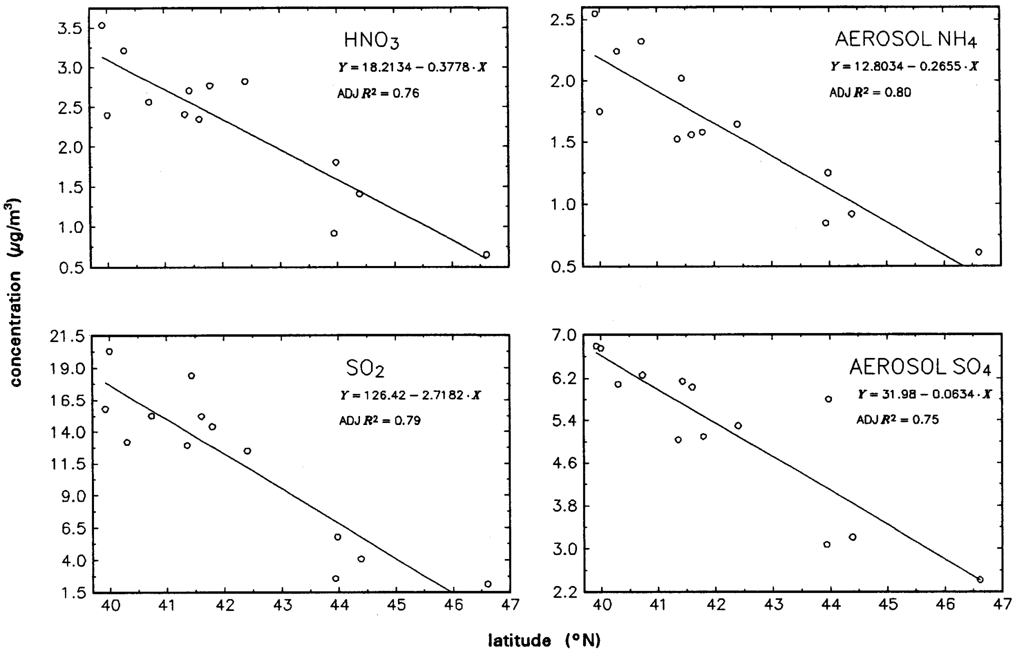


FIG. 5. Regression plots of mean annual air concentrations of dry-deposited species against latitude. Note that longitude is also a significant predictor for SO_2 and aerosol SO_4^{2-} (Table 2). No trend was observed for aerosol NO_3^- so its regional mean value (0.75 μg/m³) was used for all locations.

for NO_3^-, SO_4^{2-}, and NH_4^+ respectively, while those predicted by the model were 1.49, 2.30, and 0.240 mg/L. Lovett et al. (1982) estimated bulk deposition of SO_4^{2-} and NO_3^- to a 1220 m site on Mt. Moosilauke, New Hampshire to be 64.8 and 24.4 $kg \cdot ha^{-1} \cdot yr^{-1}$, respectively, compared with 35.9 and 22.8 $kg \cdot ha^{-1} \cdot yr^{-1}$ predicted here for wet deposition. On Madonna Mt. in Vermont (1110 m), Scherbatskoy and Bliss (1984) estimated bulk SO_4^{2-} and NO_3^- deposition to be 95.4 and 66.8 $kg \cdot ha^{-1} \cdot yr^{-1}$, respectively, whereas predicted wet deposition values for the site are 35.2 and 22.5 $kg \cdot ha^{-1} \cdot yr^{-1}$. The degree to which dry or cloud deposition may have influenced these bulk deposition measurements is unknown. The durations of these studies were also relatively short. Since deposition rates can vary greatly over short periods of time, short-term studies may not be sufficient for making accurate mean annual estimates.

Atmospheric concentrations and dry deposition

In contrast to precipitation chemistry, latitude was a more significant factor than longitude in explaining the patterns of atmospheric concentrations across the region (Fig. 5). With the exception of aerosol NO_3^-, the latitude coefficient was highly significant in all the regressions, but longitude was significant at the $P < .05$ level only for SO_2 (Table 2). Longitude was significant at the $P < .1$ level for SO_4^{2-}, and its inclusion improved the regression R^2 value, so it was retained in the final equation. The R^2 values (Table 2) indicate that the equations explain between 76 and 85% of the variance in the atmospheric concentrations of these species, except for NO_3^- particles, for which only 21% is explained and the regression is not significant.

The latitude coefficients are consistently negative, indicating that concentrations decrease from south to north. The longitude coefficients for SO_2 and SO_4^{2-} are positive, corresponding to an increase in concentrations from east to west (Table 2). Across the study area (40–47 degrees latitude, 68–80 degrees longitude), the magnitude of the latitude effect is stronger than the longitude effect for SO_2, while the two effects are more nearly equal for SO_4^{2-}. For HNO_3 and NH_4^+, the longitude coefficients were not significant and only latitude is used in the predictions.

Applying the deposition velocities discussed above (*Methods: Deposition velocities*) permits us to calculate dry deposition fluxes from the predicted concentrations. Because the deposition velocities were not measured but were estimated from literature values, these deposition fluxes are only approximations. They are meant to apply to a "typical" deciduous forest, and we assume that the deposition velocities do not change geographically within the study region. Because the aerosol NO_3^- concentrations did not show significant trends with latitude or longitude, the regional mean

TABLE 2. Regression equations describing regional patterns of air concentrations of dry deposited species (in $\mu g/m^3$, $n = 13$). All coefficients are significant at $P < .05$ except the longitude SO_4^{2-} coefficient, for which $P < .10$. Standard errors are in parentheses.

Chemical species	Coefficients			Adjusted R^2	P
	Constant	Latitude	Longitude		
SO_2	29.5814 (44.74)	−1.7584 (0.54)	0.7468 (0.32)	0.85	<.0001
Aerosol SO_4^{2-}	8.6311 (11.91)	−0.4027 (0.14)	0.1801 (0.09)	0.81	.0001
HNO_3 vapor	18.2134 (2.53)	−0.3778 (0.06)		0.76	.0001
Aerosol NO_3^-	8.7034 (8.02)	−0.1593 (0.10)	−0.0164 (0.06)	0.21	.12
Aerosol NH_4^+	12.8035 (1.62)	−0.2655 (0.04)		0.80	<.0001

value (0.75 $\mu g/m^3$) was used for all locations. In most cases, aerosol NO_3^- contributed <10% of the total dry N deposition.

Given the caveats above, predicted dry deposition of S varies from 7.5 $kg \cdot ha^{-1} \cdot yr^{-1}$ in southwestern Pennsylvania (88% as SO_2) to only 0.27 $kg \cdot ha^{-1} \cdot yr^{-1}$ in northern Maine (0% as SO_2), principally because of the strong gradient in SO_2 concentration. Predicted dry deposition of S is slightly greater than wet deposition in southwestern Pennsylvania, but is only 7% of wet deposition in northern Maine. Total dry N deposition also varies substantially across this transect, from 3.6 $kg \cdot ha^{-1} \cdot yr^{-1}$ in the southwest (78% as HNO_3, 2% as NO_3^-, and 20% as NH_4^+) to 2.2 $kg \cdot ha^{-1} \cdot yr^{-1}$ in the northeast (71% as HNO_3, 12% as NO_3^-, and 17% as NH_4^+). Predicted dry N deposition is 46% of wet deposition in southwestern Pennsylvania and 20% of wet deposition in northern Maine.

Since dry deposition was not directly measured at any of the air concentration sites and few reliable measurements have been made elsewhere in the northeast, a meaningful, region-wide validation exercise is, at present, not possible. However, dry deposition has been measured at several sites within the region using a variety of methods. Meyers et al. (1991) report rates of dry deposition of S and NO_3^{2-}-N at Whiteface Mt. and West Point, New York using an inferential method similar to the one used in the present study, but with independent concentration data and a model specifically parameterized for the sites. They estimated S dry deposition to be 2.5 and 5 $kg \cdot ha^{-1} \cdot yr^{-1}$ at Whiteface and West Point, respectively, compared to our estimates of 2.9 and 6.0 $kg \cdot ha^{-1} \cdot yr^{-1}$, respectively, for these locations. They estimated NO_3^{1-}-N dry deposition to be 1.5 and 3.0 $kg \cdot ha^{-1} \cdot yr^{-1}$, respectively, compared to our estimates of 1.4 and 2.4 $kg \cdot ha^{-1} \cdot yr^{-1}$.

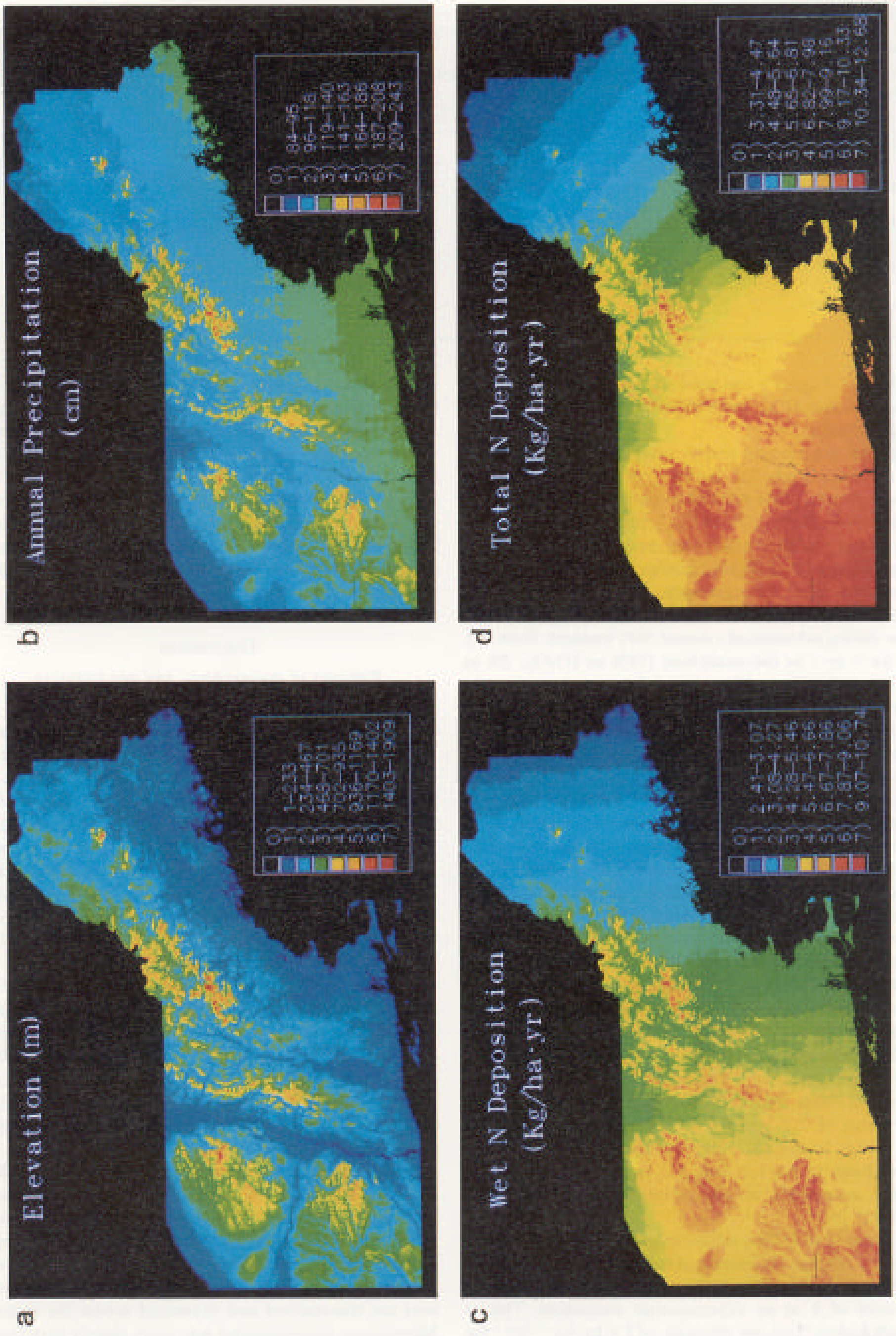
Likens et al. (1990) estimated a 23-yr average (1964–1987) S dry deposition rate of 6.7 $kg \cdot ha^{-1} \cdot yr^{-1}$ at Hubbard Brook, New Hampshire by evaluating the mass balance of S in an experimental watershed. This is much higher than our estimate of 2.6 $kg \cdot ha^{-1} \cdot yr^{-1}$ for this location. Lovett et al. (1992) compared inferential, watershed, and throughfall techniques for measuring S dry deposition at the Hubbard Brook site and found them to differ by almost a factor of 4, from 2.0 $kg \cdot ha^{-1} \cdot yr^{-1}$ for the inferential technique to 7.8 $kg \cdot ha^{-1} \cdot yr^{-1}$ for the watershed mass balance. We conclude that our estimates are reasonably close to other *inferential* estimates of dry deposition in the region, but that different methods may yield very different dry deposition flux estimates.

DISCUSSION

Patterns of atmospheric and precipitation concentrations

Precipitation and atmospheric concentrations show equally clear but distinctly different patterns across the study area. The predominant concentration trends in precipitation are the steep, west to east gradients for ions whose presence in the atmosphere is largely due to industrial activities (SO_4^{2-}, NO_3^-, and H^+). Ammonium concentration follows a similar pattern, although its main source is believed to be volatilization of NH_3 from agricultural feedlots and fertilized soils (Munger and Eisenreich 1983). The observed NH_4^+ trend could be the result of long-range transport in association with SO_4^{2-} aerosols. Hedin et al. (1990) found monthly average concentrations of NH_4^+ in bulk precipitation to be strongly correlated with SO_4^{2-} concentrations in the northeastern U.S. and Sweden. The authors attributed these trends to chemical coupling of the two ions in the atmosphere, since seasonal NH_4^+ concentrations follow SO_4^{2-} concentrations to a much greater extent than NH_3 emissions.

The east–west concentration trends of Ca^{2+} and K^+ could be explained by depletion of wind-blown soil particles from plowed fields and dirt roads in the midwestern U.S. (Munger and Eisenreich 1983), although interaction with negatively charged aerosols in the atmosphere could also influence the degree to which these ions are transported and deposited across the region. Magnesium concentration was more closely correlated


a
Elevation (m)
0
1 1–233
2 234–467
3 468–701
4 702–935
5 936–1169
6 1170–1402
7 1403–1909
b
Annual Precipitation (cm)
0
1 84–95
2 96–118
3 119–140
4 141–163
5 164–186
6 187–208
7 209–243
c
Wet N Deposition (Kg/ha·yr)
0
1 2.41–3.07
2 3.08–4.27
3 4.28–5.46
4 5.47–6.66
5 6.67–7.86
6 7.87–9.06
7 9.07–10.74
d
Total N Deposition (Kg/ha·yr)
0
1 3.31–4.47
2 4.48–5.64
3 5.65–6.81
4 6.82–7.98
5 7.99–9.16
6 9.17–10.33
7 10.34–12.68

with Na^+ and Cl^-, indicating that sea spray is the dominant source of Mg^{2+} within the region.

Contrary to precipitation concentrations, atmospheric concentrations of S and N tended to decrease from south to north, with a secondary longitude effect also present in the SO_2 and aerosol SO_4^{2-} data. This indicates that the emission sources that produce the predominant dry deposition pattern in our region are not the same as the sources that produce the predominant wet deposition pattern. The atmospheric concentrations in these studies were measured near the ground, whereas the source of ions in precipitation includes cloud condensation nuclei a thousand metres or more aloft. The high altitude material that is scavenged by precipitation is likely to have originated from more distant sources upwind, for instance the industrial areas of the Ohio River valley and the Great Lakes. Thus, it is plausible that while pollutants in precipitation show an east–west pattern, the observed patterns of dry deposition reflect closer pollutant sources, urban areas in the southern part of the study region, and show a north–south gradient. This is supported by the lack of any significant longitude effect for the atmospheric concentrations of N species, which are generally considered to be principally derived from vehicular (and thus urban) rather than industrial sources. Apparent differentiation of the chemical inputs from wet and dry deposition has also been noted in the isotope studies of Graustein and Turekian (1986), and Lovett and Lindberg (*in press*) noted a lack of correlation between air and precipitation concentrations of nitrogen species in a number of study sites in North America.

The data also indicate that dry-deposited species, particularly the gaseous species (SO_2 and HNO_3), decrease more rapidly with distance from source areas than do precipitation-borne oxides of S and N. For example, as one moves from southwest to northeast across the study area, sulfur dioxide concentrations decrease from about 19 $\mu g/m^3$ to near 0 and HNO_3 concentrations decrease by a factor of 5. These trends suggest that dry deposition may be a more near-source phenomenon than wet deposition.

Regional patterns of total deposition

Combining the east–west wet deposition gradients of N and S compounds with the predominantly north–south dry deposition gradients produces trends of decreasing total deposition from the southwest to the northeast within the region. Although some uncertainty exists regarding the magnitude of dry deposition estimates, the relative spatial patterns observed should be more reliable because broad-scale dry deposition patterns are more likely to result from air concentration trends than regional changes in deposition velocities.

Using the deposition velocity estimates discussed above (*Methods: Deposition velocities*), predicted total deposition of sulfur decreases from about 19 $kg \cdot ha^{-1} \cdot yr^{-1}$ in southwestern Pennsylvania (40° N, 80° W, 300 m elevation) to 5 $kg \cdot ha^{-1} \cdot yr^{-1}$ in northern Maine (46° N, 69° W, 300 m). Predicted total nitrogen deposition decreases from about 11.5 $kg \cdot ha^{-1} \cdot yr^{-1}$ to 5 $kg \cdot ha^{-1} \cdot yr^{-1}$ along the same gradient. Fig. 6 illustrates the steps by which N deposition fields for the eastern half of the region are produced in the GIS. First, the digital elevation model (Fig. 6a) is combined with Eqs. 1 and 2 to determine estimated precipitation amounts (Fig. 6b). Then, the regional wet N concentration trends (Table 1) are combined with the precipitation field to produce a map of wet N deposition (Fig. 6c). Finally, regional trends in predicted dry N deposition are added to produce a map of total N deposition (Fig. 6d).

Not included in the deposition estimates are cloud water inputs, nor was any attempt made to include elevational trends in dry deposition. It should also be remembered that all of the measurement sites for air and precipitation chemistry used in this study are located in rural areas to avoid local enhancement of atmospheric deposition from cities and other large pollution sources. Although such sources exist within the study region, especially in southern and coastal areas, their local effects are not accounted for in the deposition patterns shown here, because necessary data are not available to quantify them.

The regional deposition gradients observed for S, N and H^+ may have important ecological and environmental ramifications. If atmospheric inputs play a strong role in the function of ecosystems, we might expect these deposition patterns to be reflected eventually in regional biogeochemistry and ecosystem health. Several reports have suggested that such signals may already exist. McNulty et al. (1990) found increases in soil N content and nitrification potential in spruce-fir forests along a transect from Maine to New York. Recent studies summarizing surface water chemistry in New York state show highest and increasing levels of NO_3^- in the Catskill Mountain area with lesser increases in the Adirondacks (Driscoll et al. 1989, Murdoch and Stoddard 1992). Within the Adirondacks, Driscoll and Van Dreason (*in press*) have identified a

←

FIG. 6. Generation of a nitrogen deposition map within a geographic information system. (a) The digital elevation model (DEM) of eastern New York and New England. (b) The regional precipitation field, created by the interaction between the DEM and precipitation equations (1 and 2). (c) Predicted wet deposition of nitrogen (as elemental N) for the region, generated by combining the regional N concentration gradient (from NO_3^- and NH_4^+, Table 1) with the regional precipitation field. (d) Predicted total N deposition for the region, generated by combining equations for dry N deposition (from Table 2 and deposition velocity estimates) with the regional wet N deposition field.

trend of increasing NO_3^- concentration to the west and south, corresponding to the N deposition gradient. The few data available suggest that surface water NO_3^- concentrations across the eastern end of the northeast region remain relatively low (Driscoll et al. 1989, Stoddard, *in press*).

Other possible effects of these deposition patterns include influences on forest health and productivity. Although there is no general agreement on causes of the observed decline of certain high-elevation spruce forests in the region, it is interesting to note that the decline appears to follow patterns of deposition, being greatest in the Adirondack Mountains of New York and decreasing towards the east through Vermont, New Hampshire, and Maine (Vogelmann and Rock 1988, Craig and Friedland 1990).

Elevation effects and cloud water deposition

The elevational increase in deposition included in the present model is a function of increases in predicted precipitation amounts only. Confidence in this relationship is limited by the scarcity of available information regarding deposition to high-elevation sites. First, surprisingly few long-term precipitation collection stations have been established at high elevations, forcing the orographic precipitation effects derived here to be heavily influenced by only a few data points. Second, we have assumed that there is no change in precipitation chemistry with elevation, based on several site-specific studies that examined elevation differences of as much as 1500 m in the eastern United States (Tennessee Valley Authority 1983, Scherbatskoy and Bliss 1984, Lindberg et al. 1988). Although data from the present study show a decrease in concentration with elevation for most ions, we maintained the assumption of constant concentration at all elevations due to the limited elevation range of the data and the seemingly unrealistic coefficients obtained. Additionally, comparison of model predictions with data collected by Miller et al. (*in press*) at 1050 m on Whiteface Mt., New York showed no change in concentration with elevation for NO_3^- and NH_4^+, although the concentration of SO_4^{2-} was slightly lower than that predicted. More work is clearly needed to establish confidently the relationship between elevation and ion concentrations in precipitation.

Elevation effects on dry deposition could not be included in the model since so little relevant information exists. Some increase in dry deposition input may be expected with increasing elevation, however, as a result of several factors. In the northeast, high elevation sites tend to be windier, wetter, and contain a greater proportion of needle-bearing trees than adjacent low-elevation sites, which may combine to enhance rates of delivery and adhesion of suspended particles and gases to canopy surfaces (Lovett and Kinsman 1990).

Also absent from the model are cloud water inputs. Cloud water can contain very high concentrations of dissolved ions and can constitute a large portion of total deposition to high-elevation sites, which are frequently immersed in clouds (Lovett et al. 1982, Scherbatskoy and Bliss 1984, Weathers et al. 1988, Lovett and Kinsman 1990, Vong et al. 1991, Miller et al., *in press*). Although too few data exist to conduct a region-wide spatial analysis of cloud deposition, it might be possible to derive estimates as a function of wet deposition and elevation. Lovett and Kinsman (1990) presented a relationship between elevation and percent of total SO_4^{2-} deposition contributed by cloud water. In the northeastern U.S., cloud deposition appears to begin at ≈600 m and increases at a rate of 0.094% (of total deposition) per metre. Most of the values used to obtain this relationship, however, did not include dry deposition and those that did reported only small contributions, so it may be possible to estimate cloud input of SO_4^{2-} for sites above 600 m with an equation such as the following:

$$SO_4{}^{2-}{}_{\text{cloud}} = \frac{SO_4{}^{2-}{}_{\text{wet}}}{1.564 - (\text{elev}\cdot 0.00094)} - SO_4{}^{2-}{}_{\text{wet}}, \quad (3)$$

where $1.564 - (\text{elev}\cdot 0.00094)$ is the proportion of total deposition contributed by precipitation.

Such an approach could provide a useful method of adding a cloud deposition component to the regional model, although longer term studies are needed before such relationships can be considered reliable.

Conclusion

The processes that affect atmospheric deposition can be extremely variable across time and space. Over short periods of time, factors such as precipitation events may appear highly random, making spatial patterns difficult to discern. After long-term averages of deposition variables are obtained, however, statistically meaningful trends can be confidently established.

This study has identified patterns of both wet and dry deposition within the northeastern United States that have important implications for regional biogeochemistry and ecosystem health. However, it has also been instructive in pointing out weaknesses in existing data. For improved understanding of atmospheric inputs to ecosystems, increased attention should be given to devising standard methods for measuring dry and cloud water deposition, and monitoring these processes across various spatial scales. Greater attention should also be given to monitoring deposition at high elevations and near urban areas where deposition rates are likely to be elevated. Although we have emphasized broad-scale deposition trends, fine-scale features such as local wind exposure and vegetation type are also likely to affect deposition inputs (Weathers et al. 1992). Field studies designed to detect variation at this level are also warranted.

Acknowledgments

We gratefully acknowledge the efforts of J. Bognar who aided with the GIS data processing. We also thank L. Pitelka, D. Raynal, and an anonymous reviewer for making valuable comments on an earlier version of this manuscript. Funding for this project was provided by the USDA Forest Service Northeastern Experiment Station (Co-op Agreement #23-605), the National Aeronautics and Space Administration (Grant #NAGW 1825) and the National Science Foundation (Grant #BSR-8910988 and through the Long Term Ecological Research Program-Hubbard Brook site). Additional funding was provided by the General Reinsurance Corporation through a grant to the Institute of Ecosystem Studies. This is a contribution to the Hubbard Brook Ecosystem Study and the program of the Institute of Ecosystem Studies.

Literature Cited

Aber, J. D., K. J. Nadelhoffer, P. Steudler, and J. M. Melillo. 1989. Nitrogen saturation in northern forest ecosystems—hypotheses and implications. BioScience **39**:378–386.

Barrie, R. G. 1981. Mountain weather and climate. Methuen, New York, New York, USA.

Craig, B. W., and A. J. Friedland. 1990. Spatial patterns in forest composition and standing dead red spruce in montane forests of the Adirondacks and northern Appalachians. Environmental Monitoring and Assessment **18**:129–143.

Cronan, C. S., and C. L. Schofield. 1980. Aluminum leachate response to acid precipitation: effects on high-elevation watersheds in the northeast. Science **204**:304–306.

Driscoll, C. T., D. A. Schaefer, L. A. Malot, and P. J. Dillon. 1989. Summary of North American data. Page 10 *in* J. L. Malanchuk and J. Nilsson, editors. The role of nitrogen in the acidification of soils and surface waters. Nordic Council of Ministers, Miljorapport, Stockholm, Sweden.

Driscoll, C. T., and R. Van Dreason. *In press.* Seasonal and long-term patterns in the chemistry of Adirondack lakes. Water, Air and Soil Pollution.

Edgerton, E. S., and T. F. Lavery. 1991. National Dry Deposition Network: Fourth Annual Progress Report (1990). Report of contract no. 68-02-4451. U.S. Environmental Protection Agency, Research Triangle Park, North Carolina.

Edgerton, E. S., T. F. Lavery, and H. S. Prentice. 1991. National Dry Deposition Network: Third Annual Progress Report (1989). EPA/600/3-91/018, U.S. Environmental Protection Agency, Office of Research and Development, Washington, D.C., USA.

Federer, C. A., L. D. Flynn, C. W. Martin, J. W. Hornbeck, and R. S. Pierce. 1990. Thirty years of hydrometeorologic data at the Hubbard Brook Experimental Forest, New Hampshire. General Technical Report NE-141. U.S. Department of Agriculture, Forest Service, Northeastern Forest Experimental Station, Radnor, Pennsylvania, USA.

Fowler, D., J. N. Cape, I. D. Leith, I. W. Choularton, M. J. Gray, and A. Jones. 1988. The influence of altitude on rainfall composition at Great Dun Fell. Atmospheric Environment **22**:1355–1362.

Friedland, A. J., R. A. Gregory, L. Karenlampi, and A. H. Johnson. 1984. Winter damage as a factor in red spruce decline. Canadian Journal of Forest Research **14**:963–965.

Graustein, W. C., and K. K. Turekian. 1986. 210-Pb and 137-Cs in air and soils measure the rate and vertical profile of aerosol scavenging. Journal of Geophysical Research **91**(D13):14355–14366.

Hamilton, L. C. 1990. Modern data analysis: a first course in applied statistics. Brooks/Cole, Pacific Grove, California, USA.

Hanson, P. J., K. Rott, G. E. Taylor, C. A. Gunderson, S. E. Lindberg, and B. M. Ross-Todd. 1989. NO_2 deposition to elements representative of a forest landscape. Atmospheric Environment **23**:1783–1794.

Hedin, L. O., L. Granat, G. E. Likens, and H. Rodhe. 1990. Strong similarities in the seasonal concentration ratios of SO_4^{-2}, NO_3^-, and NH_4^+ in precipitation between Sweden and the northeastern U.S. Tellus **42B**:454–462.

Hedin, L. O., G. E. Likens, and F. H. Bormann. 1987. Decrease in precipitation acidity resulting from decreased SO_4^{2-} concentration. Nature **325**:244–246.

IES [Institute of Ecosystem Studies]. 1988, 1989, 1990. Annual air quality report—1988, 1989, 1990. Institute of Ecosystem Studies, Milbrook, New York, USA.

Johnson, D. W., and S. E. Lindberg, editors. 1992. Atmospheric deposition and forest nutrient cycling. Springer-Verlag, New York, New York, USA.

Likens, G. E., F. H. Borman, L. O. Hedin, C. T. Driscoll, and J. S. Eaton. 1990. Dry deposition of sulfur: a 23 year record for the Hubbard Brook Forest Ecosystem. Tellus **42B**:319–329.

Lindberg, S. E., G. M. Lovett, D. D. Richter, and D. W. Johnson. 1986. Atmospheric deposition and canopy interactions of major ions in a forest. Science **231**:93–102.

Lindberg, S. E., D. Silsbee, D. A. Schaefer, J. G. Owens, and W. Petty. 1988. A comparison of atmospheric exposure conditions at high- and low-elevation forests in the southern Appalachian Mountain Range. Pages 321–344 *in* M. H. Unsworth and D. Fowler, editors. Deposition processes at high elevation sites. Kluwer, Dordrecht, The Netherlands.

Lovett, G. M., and J. D. Kinsman. 1990. Atmospheric pollutant deposition to high elevation ecosystems. Atmospheric Environment **24**:2767–2786.

Lovett, G. M., G. E. Likens, and S. S. Nolan. 1992. Dry deposition of sulfur to the Hubbard Brook Experimental Forest: a preliminary comparison of methods. Pages 1391–1400 *in* S. E. Schwartz and W. G. N. Slinn, editors. Precipitation scavenging and atmosphere-surface exchange. Volume 3. The Summers volume: application and appraisals. Hemisphere Publishing, Washington, D.C., USA.

Lovett, G. M., and S. E. Lindberg. *In press.* Atmospheric deposition and canopy exchange of nitrogen in forests. Canadian Journal of Forest Research.

Lovett, G. M., W. A. Reiners, and R. K. Olsen. 1982. Cloud droplet deposition in subalpine balsam-fir forests: hydrological and chemical inputs. Science **218**:1303–1304.

McNulty, S. G., J. D. Aber, T. M. McLellan, and S. M. Katt. 1990. Nitrogen cycling in high-elevation forests in the northeastern U.S. in relation to nitrogen deposition. Ambio **19**:38–40.

Meyers, T. P., B. B. Hicks, R. P. Hosker Jr., J. D. Womack, and L. C. Satterfield. 1991. Dry deposition inferential measurement techniques—II. Seasonal and annual deposition rates of sulfur and nitrate. Atmospheric Environment **25A**:2361–2370.

Miller, E. K., J. A. Panek, A. J. Friedland, J. Kadlecek, and V. A. Mohnen. *In press.* Atmospheric deposition to a high-elevation forest at Whiteface Mountain, New York, USA. Tellus.

Munger, J. W., and S. J. Eisenreich. 1983. Continental-scale variation in precipitation chemistry. Environmental Science and Technology **17**:32a–42a.

Murdoch, P. S., and J. L. Stoddard. 1992. The role of nitrate in the acidification of streams in the Catskill Mountains of New York. Water Resources Research **28**:2707–2720.

NADP/NTN [National Atmospheric Deposition Program (IR-7)/National Trends Network]. 1/23/1991. NADP/NTN Coordinating Office, Natural Resource Ecology Laboratory, Colorado State University, Fort Collins, Colorado, USA.

Nihlgard, B. 1985. The ammonium hypothesis—an additional explanation to the forest decline in Europe. Ambio **14**:2–8.

NOAA [National Oceanic and Atmospheric Administration]. 1982. Climatography of the United States Number 81 (by state): monthly normals of temperature, precipitation, and heating and cooling degree days 1951–1980. NOAA Environmental Data and Information Service, National Climatic Center, Asheville, North Carolina, USA.

Reuss, J. O. and D. W. Johnson. 1986. Acid deposition and the acidification of soils and waters. Springer-Verlag, New York, New York, USA.

Scherbatskoy, T., and, M. Bliss. 1984. Occurrence of acid rain and cloudwater in high elevation ecosystems in the Green Mountains, Vermont. Pages 449–463 *in* P. J. Sampson, editor. The meteorology of acid deposition. Air Pollution Control Association, Pittsburgh, Pennsylvania, USA.

Schulze, E.-D. 1989. Air pollution and forest decline in a spruce (*Picea abies*) forest. Science **224**:776–783.

Stoddard, J. L. *In press.* Long-term changes in watershed retention of nitrogen: its causes and aquatic consequences. *In* L. A. Baker, editor. Environmental chemistry of lakes and reservoirs. Progress in Chemistry Series. American Chemical Society, Washington, D.C., USA.

Summers, P. W., V. C. Brewersox, and G. J. Stensland. 1986. The geographical distribution and temporal variation of acidic deposition in eastern North America. Water, Air and Soil Pollution **31**:523–535.

Tennessee Valley Authority. 1983. Investigations of the cause of fishkills in fish-rearing facilities in Raven Fork watershed. TVA/ONR/WR-83/9, Tennessee Valley Authority, Chattanooga, Tennessee, USA.

United States Geologic Survey. 1987. Digital elevation models: data user's guide 5. U.S. Geologic Survey, Reston, Virginia, USA.

van Breeman, N., and H. F. G. van Dijk. 1988. Ecosystem effects of atmospheric deposition of nitrogen in the Netherlands. Environmental Pollution **54**:249–274.

Vogelmann, J. E., and B. N. Rock. 1988. Assessing forest damage in high-elevation coniferous forests in Vermont and New Hampshire using Thematic Mapper data. Remote Sensing of Environment **24**:227–246.

Vong, R. J., Sigmund, J. T., and S. F. Mueller. 1991. Cloud water deposition to appalachian forests. Environmental Science and Technology **25**:1014–1021.

Weathers, K. C., G. E. Likens, F. H. Borman, S. H. Bicknell, B. T. Borman, B. C. Daube Jr., J. S. Eaton, J. N. Galloway, W. C. Keene, K. D. Kimball, W. H. McDowell, T. G. Siccama, D. Smiley, and R. A. Tarrant. 1988. Cloud water chemistry from ten sites in North America. Environmental Science and Technology **22**:1018–1026.

Weathers, K. C., G. M. Lovett, and G. E. Likens. 1992. The influence of a forest edge on cloud deposition. Pages 1415–1423 *in* S. E. Schwartz and W. G. N. Slinn, editors. Precipitation scavenging and atmosphere-surface exchange. Volume 3. The Summers volume: application and appraisals. Hemisphere Publishing, Washington, D.C., USA.

Westerfelt, J., W. Goran, and M. Shapiro. 1987. Design and development of GRASS: the Geographic Resources Analysis Support System. U.S. Army Construction Engineering Research Laboratory, Champaign, Illinois, USA.

Wilson, J. W., and V. A. Mohnen. 1982. An analysis of spatial variability of the dominant ions in precipitation in the eastern United States. Water, Air and Soil Pollution **18**: 199–213.

Wisniewski, J., and J. D. Kinsman. 1988. pH and hydrogen ion deposition patterns for the continental United States and Canada. Water, Air and Soil Pollution **38**:1–17.

Zemba, S. G., D. Golomb, and J. A. Fay. 1988. Wet sulfate and nitrate deposition patterns in eastern North America. Atmospheric Environment **22**:2751–2761.