EMPIRICAL MODELING OF ATMOSPHERIC DEPOSITION IN MOUNTAINOUS LANDSCAPES

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Abstract. Atmospheric deposition has long been recognized as an important source of pollutants and nutrients to ecosystems. The need for reliable, spatially explicit estimates of total atmospheric deposition (wet + dry + cloud) is central, not only to air pollution effects researchers, but also for calculation of input–output budgets, and to decision makers faced with the challenge of assessing the efficacy of policy initiatives related to deposition. Although atmospheric deposition continues to represent a critical environmental and scientific issue, current estimates of total deposition have large uncertainties, particularly across heterogeneous landscapes such as montane regions.

We developed an empirical modeling approach that predicts total deposition as a function of landscape features. We measured indices of total deposition to the landscapes of Acadia (121 km²) and Great Smoky Mountains (2074 km²) National Parks (USA). Using ~300–400 point measurements and corresponding landscape variables at each park, we constructed a statistical (general linear) model relating the deposition index to landscape variables measured in the field. The deposition indices ranged over an order of magnitude, and in response to vegetation type and elevation, which together explained ~40% of the variation in deposition. Then, using the independent landscape variables available in GIS data layers, we created a GIS-relevant statistical nitrogen (N) and sulfur (S) deposition model (LandMod). We applied this model to create park-wide maps of total deposition that were scaled to wet and dry deposition data from the closest national network monitoring stations. The resultant deposition maps showed high spatial heterogeneity and a four- to sixfold variation in “hot spots” and “cold spots” of N and S deposition ranging from 3 to 31 kg N ha⁻¹ yr⁻¹ and from 5 to 42 kg S ha⁻¹ yr⁻¹ across these park landscapes. Area-weighted deposition was found to be up to 70% greater than NADP plus CASTNET monitoring-station estimates together. Model-validation results suggest that the model slightly overestimates deposition for deciduous and coniferous forests at low elevation and underestimates deposition for high-elevation coniferous forests. The spatially explicit deposition estimates derived from LandMod are an improvement over what is currently available. Future research should test LandMod in other mountainous environments and refine it to account for (currently) unexplained variation in deposition.

Key words: Acadia National Park (USA); atmospheric deposition; elevation; GIS; Great Smoky Mountain National Park (USA); hotspots; landscape features; model; nitrogen; sulfur; throughfall; vegetation type.

INTRODUCTION

Atmospheric deposition is an important source of nutrients and pollutants to ecosystems. For many terrestrial ecosystems it is the primary source of nitrogen and sulfur (Likens and Bormann 1995, Schlesinger 1997). Accurate measures of total atmospheric deposition are critically important for many reasons, including the calculation of input–output budgets, assessment of biological, ecological, and watershed responses to pollutant and nutrient loading, and for linking air emissions with actual deposition to landscapes (Weatherers and Lovett 1998, Driscoll et al. 2001, Holland et al. 2005). For example, ecosystem theory and mechanisms of nutrient cycling have been proposed based on budget imbalances: the difference between watershed outputs and inputs has been used in support of biogeochemical “theory” that suggests that old-growth, temperate forests are less retentive of nitrogen than vigorously growing or disturbed watersheds (Vitousek and Reiners 1975, Hedin et al. 1995, Goode et al. 2000, Aber et al. 2003). In addition, for much of Europe, deposition estimates are critical for setting emission-control policies and legislation (Hettelingh et al. 1995a, b).

Substances are delivered from the atmosphere to Earth’s surface in three forms: wet deposition (rain and snow), dry deposition (gases and particles), and cloud or
fog (occult) deposition (Lovett 1994). We know much about the contribution of wet deposition to nutrient and pollutant budgets, but relatively little about dry and cloud deposition (Weathers et al. 2000).

**Wet deposition**

As a result of intensive, site-specific, long-term monitoring efforts (Likens and Bormann 1995, Kelly et al. 2002) as well as the existence of the national monitoring networks, both wet-chemistry and deposition data are readily available from more than 250 monitoring stations in the United States (National Atmospheric Deposition Program [NADP; information available online]$^5$ and AIRMoN [information available online]$^6$). Wet deposition is the product of precipitation amount and chemistry, so point estimates can also be extended with some accuracy to weather stations where only precipitation amount is monitored. Thus temporal trends (since the late 1970s) and spatial distribution of wet deposition are well documented in the United States and Canada and Europe (Hedin et al. 1994, Summers 1995, Lynch 2004, Holland et al. 2005) and in many parts of Canada and Europe (Hedin et al. 1994, Summers 1995, van Leeuwen et al. 1996, Holland et al. 2005).

**Dry deposition**

Estimates of dry deposition are far less certain than wet deposition, both because of the paucity of air-chemistry monitoring stations and the inadequacy of the models used for estimating dry deposition. Air chemistry is measured routinely at independent research sites (Kelly et al. 2002) as well as nationally through the Clean Air Status and Trends Network (CASTNET; information available online)$^5$ and AIRMoN (see footnote 6) monitoring programs. There are relatively few air-chemistry monitoring stations in the United States (CASTNET [see footnote 7] and AIRMoN [see footnote 6]), with most of these located in the eastern half of the country. Most estimates of dry-particulate and gas deposition are based on measured air concentrations, which then are used in dry-deposition models (Meyers et al. 1998, Finkelstein et al. 2000) that consider the multiple resistances, canopy characteristics, and meteorological variables that influence deposition (see Lovett 1994). Current dry-deposition models are limited by simplifying assumptions, which include homogeneous canopies and flat terrain (Weathers et al. 1995, 2000). Dry-deposition model estimates are reasonably robust in grasslands and croplands with flat terrain, but perform more poorly in forests, and are largely untested in montane and coastal regions. However, cloud chemistry measurements and monitoring programs are few. Cloud chemistry data for the United States have been collected from fewer than 20 mountains and even fewer Atlantic and Pacific coastal areas (Weathers et al. 1986, 1988, Kimball et al. 1988, Vong et al. 1991, Mohlen and Vong 1993, Anderson et al. 1999, Baumgardner et al. 2003). Cloud-deposition models suffer from the same limitations as dry-deposition models in that they rely on cloud chemistry data from single stations, and on measured or modeled meteorologic and canopy variables, and do not account for complex topography or heterogeneous vegetation (Weathers et al. 2000).

**Total deposition**

Total deposition (wet + dry + cloud) estimates for complex terrain are either inadequate or lacking, primarily as a result of the vagaries of measuring dry, cloud, and/or snow deposition in complex terrain. A sulfur- and nitrogen-deposition map was created in the early 1990s for a limited area of the northeastern United States (Ollinger et al. 1993). It has been extensively used by the research community to determine site-specific deposition estimates, even though it does not have sufficient resolution to accurately discriminate between locations that are 10s of kilometers apart. This map does not include cloud deposition, and its dry-deposition estimates do not take into account elevational effects or variations in land cover, reducing its accuracy in complex terrain. In recent years, other important deposition maps have been published for the United States (Holland et al. 2005) and limited areas in the western United States (Fenn et al. 2003, Nanus et al. 2003). Despite these efforts, no empirical model exists for total deposition to heterogeneous terrain.

**Objectives**

We developed an empirical modeling approach to explore controls on deposition across heterogeneous landscapes. The specific goals of this work were: (1) to develop a generalizable method whereby both total deposition to heterogeneous terrain and its spatial heterogeneity across a landscape could be more accurately measured; (2) to determine which readily measured field- and GIS-derived independent variables control deposition rate in an empirically based statistical model; and (3) to develop landscape-scale maps of nitrogen (N) and sulfur (S) deposition (LandMod) from dry- and wet-deposition monitoring data (“reference deposition”) and a separate empirically based statistical model using only independent variables that were available as geographic information system (GIS) data layers. Finally, (4) we examined the behavior of our model by performing validation analyses.
METHODS

General approach

We measured indices of total deposition in the field and converted those measures to unitless “scaling factors” by relating them to total deposition from a low-elevation, flat area. Using the scaling factors, we developed two statistical models, one that used independent variables measured in the field (e.g., elevation, canopy type, diameter at breast height, canopy cover), “StatMod,” and one that used a subset of these variables that were available as data layers in a GIS (e.g., aspect, elevation, vegetation type). We used the latter, “LandMod,” plus monitoring data, in a GIS to create maps of deposition for landscapes. StatMod identified which independent variables, out of all available, control atmospheric deposition in mountainous landscapes, while LandMod (a combination GIS–statistical model) allowed scaling deposition estimates over an entire large area. Throughout the Methods section we refer to Fig. 1, which outlines the complete procedure, from collecting data in the field to creating deposition maps; this is our process for scaling up from point measurements to the landscape.

Study areas

We conducted our research in Acadia National Park (ACAD) and in Great Smoky Mountains National Park (GRSM). Atmospheric deposition of pollutants has been identified as a primary threat to natural resources in both of these parks by the National Park Service, and both parks have wet and dry atmospheric deposition monitoring stations. ACAD is located on the north Atlantic coast of the United States in Maine (see Plate 1). The ACAD study area of interest was the entire Mount Desert Island (278 km²). GRSM is located in the southern Appalachian Mountains of the United States, in Tennessee and North Carolina. GRSM has a total area of 2074 km², and is ~450 km from the nearest coast.

GIS data acquisition

Vegetation and elevation (DEM [digital elevation model]) data layers were acquired for both ACAD and GRSM. For each park we used a USGS 1:24000 DEM with 30 × 30 m pixels. Vegetation for GRSM was derived from LandSat Thematic Mapper imagery collected in September 1984 (MacKenzie 1993). Vegetation data for ACAD, derived from 1:15840 color
infrared aerial photographs collected 27–28 May 1997, were used (Lubinski et al. 2003). Vegetation data were reclassified as conifer forest, deciduous forest, mixed conifer–deciduous forest, or nonforest and summarized in the “study area” sections of Table 1a (ACAD) and Table 1b (GRSM). Elevation data (meters above sea level) were used to calculate slope (degrees) and aspect (degrees) data layers for each park, using a 3 × 3 pixel moving window (Table 1). Elevation data were also used to calculate a unitless index of topographic exposure, or landscape position, that specifies whether a location is on an exposed ridge (negative values), a flat area (values at or near zero), or in a depression (positive values).

**Field methods for Acadia National Park**

The deposition index (DI; Fig. 1) for ACAD was sulfur in throughfall (TF). Throughfall S has been shown to be a conservative tracer of wet, dry, and cloud deposition for forests of the eastern United States (see Discussion: Indices of deposition, below, and Lindberg and Lovett [1992], Weathers et al. [1992, 1995], Lovett [1994]). Two hundred eighty-five resin throughfall collectors (Simkin et al. 2004) were installed on National Park Service property along a set of trail loops that could be accessed in 1–2 days to minimize the risk of precipitation events interrupting the sample collection. Each throughfall collector was located halfway between the trunk and drip line of a tree. Five resin bulk-collector locations were also installed in the open: one at 413-m elevation on Cadillac Mountain (see Plate 1), one at 405-m elevation on Sargent Mountain, one at 17-m elevation within 200 m of the coast, and two at 153–157 m elevations in the same clearing as the permanent monitoring stations. The resin throughfall (and resin bulk collector) sampling season was from 7 June 2000 to 18 September 2000, subdivided into three sampling periods of 4–6 weeks each.

At each collector vegetation and terrain attributes were recorded, as well as position coordinates. Forest type, tree species name, tree diameter at breast height, tree height, qualitative tree-canopy density, and understory type were recorded for vegetation in the area of canopy influence (assumed to be an inverted cone extending upward from the collector at 45 degrees). A compass and a clinometer were used to record the aspect and slope, respectively, of the terrain immediately downslope of the collector. A digital hemispherical photograph was taken during midday hours under cloudy conditions to quantify tree-canopy closure. Position coordinates were recorded with a GPS unit, with the resulting horizontal precision ranging from 0.148 to 2.343 m. Vegetation, elevation, slope, and aspect attributes of collectors were also extracted from GIS data layers using GPS coordinates, and are summarized in the sampling points section of Table 1a.
Field Methods for Great Smoky Mountains National Park

The deposition index (DI; Fig. 1) for GRSM was the concentration of lead (Pb) in the soil organic horizon. It was impractical to measure spatially representative TF in an area as large as GRSM. Lead has been shown to preserve the pattern of long-term total deposition (see Discussion: Indices of deposition, below, and Johnson et al. [1982], Weathers et al. [1995, 2000]). As a test of this method, we previously measured Pb concentrations in forest-floor samples collected at seven NADP sites along a regional deposition gradient in the northeastern United States (Lovett and Rueth 1999). There was a significant positive, linear relationship for Pb (in milligrams per kilogram) with both total (wet + dry) S ($r^2 = 0.95, n = 91$ samples) and total (wet + dry) inorganic N ($r^2 = 0.72, n = 91$ samples) deposition, demonstrating that Pb in the forest floor reflects patterns of deposition across latitudinal, temperature, and 3–4-fold depositional gradients spanning from Pennsylvania to Maine.

We collected Pb samples from 378 forest-floor locations in GRSM during 1999 and 2000. Sampling was constrained to a ~800-km$^2$ rectangular swath (22 km wide and 37 km long; oriented diagonally from northwest to southeast; northwest corner 35.69° N and 83.65° W; southeast corner 35.54° N and 83.23° W) that passes through the center of the park. Samples were collected >100 m from primary roads and were 15–25 m upslope of trails. Areas with evidence of fire or organic horizon disturbance were avoided. Potential sampling areas were also rejected if they did not have an intact forest canopy (>12 m tall or >25-cm-diameter trees) that would have collected Pb during most if not all of the 50–60 years preceding sample collection. An additional six samples from a deciduous forest adjacent to the NADP site (TN11) were collected for use as an average Pb base deposition value (BD, Fig. 1).

At the area to be sampled, a centerpoint was established and two subsamples of the organic horizon were collected in each cardinal direction, for a minimum of 8 subsamples. If the organic horizon was thin with insufficient sample volume, additional subsamples were collected (up to 16 subsamples) within the 5-m-radius sampling area. The subsamples of the organic horizon (up to ~9 cm deep) were collected using a soil corer with a diameter of 5.77 cm. Subsamples were composited into a single sample.

Vegetation and terrain attributes, as well as position coordinates, were recorded at each sampling area. Forest type, trees species names, qualitative tree canopy cover, understory type, slope and aspect were all measured in the field. The coordinates of the centerpoint of the organic matter sampling area were recorded with a GPS unit, with the resulting proprietary figure-of-merit (FOM) accuracy values ranging from 4.6 to 37.5 m. Vegetation, elevation, slope, and aspect attributes of the 378 GRSM organic-horizon sampling locations were also extracted from GIS data layers using GPS coordinates, and are summarized in the “sampling points” section of Table 1b.

During the second year of the study, throughfall samples were collected, as described above, from 32 locations and were used to validate our LandMod model.

Laboratory methods

The resin throughfall columns were extracted in 1.0 mol/L potassium iodide (KI) and analyzed for sulfate ($\text{SO}_4^{2-}$) with a Dionex ion chromatograph as described in Simkin et al. (2004). Three resin column “blank” samples were extracted along with the field resin samples for each sample period. Raw $\text{SO}_4^{2-}$ concentrations were converted to S fluxes (in kilograms per hectare per sampling period) by taking into account the collection area and the period of time that resin columns were exposed in the field.

Forest-floor samples were dried, sieved, ground, and digested after the methods of Weathers et al. (1995, 2000). A laboratory blank (empty crucible) and sample of standard reference material (NIST [National Institute of Standards and Technology] 2781, domestic sludge) were included with each extraction batch. Digested material was filtered through number 42 ashless Whatman paper and brought to volume with double-deionized water, and then analyzed for Pb using an inductively coupled plasma atomic-emission spectrometer. Raw laboratory-sample Pb concentrations (in milligrams per liter) were converted to surface organic-horizon concentrations (milligrams of Pb per kilogram).

For both methods, NIST-traceable knowns, standards-as-samples, and replicate samples were included in each instrument run for quality-control purposes. The quality assurance–quality control (QA/QC) results for both methods were within ±10% accuracy and precision.

Calculation of base deposition (BD) and scaling factors (SF)

Base deposition (BD; Fig. 1) was the deposition-index measurement at a low-elevation site during the time period that the field sampling occurred. At ACAD the BD was calculated by summing weekly NADP wet deposition and CASTNET dry deposition of S (in kilograms per hectare) from the ACAD monitoring station for the period (6 June 2000 to 19 September 2000) that most closely matched the throughfall sampling period. At GRSM the BD was calculated from the mean Pb concentration (in milligrams per kilogram) of six organic-horizon samples collected in a forest adjacent to the NADP monitoring location.

Deposition indices (kilograms S per hectare in throughfall for ACAD and milligrams Pb per kilogram in organic horizon for GRSM) were then divided by respective BDs to produce unitless deposition scaling factors (SFs) (Fig. 1). SFs for the five nonforested collectors at ACAD were calculated by dividing by NADP wet deposition only. It was not possible to
calculate SFs for nonforested areas at GRSM since it was not possible to find a range of nonforested elevations from which we could collect undisturbed organic-horizon samples.

Reference deposition

Reference deposition (RD) was annual wet + dry deposition measured at NADP and CASTNET monitoring stations (Fig. 1). These stations were ME98 (NADP wet) and ACA416 (CASTNET dry) for ACAD (44.3739° N and 68.2606° W) and TN11 (NADP wet; 35.6645° N and 83.5903° W) and GRS420 (CASTNET dry; 35.6331° N and 83.9422° W) for GRSM. We used the data for calendar year 2000 as a representative year and scaled up from point measurements to create spatially explicit deposition maps for each of the parks, described below (see Results: Statistical model results: Modeled N and S deposition maps: LandMod).

Statistical analyses

Our first goal was to create an empirically based statistical model of measured deposition as a function of field and GIS variables (StatMod). All statistical analyses were conducted using the SAS statistical package (SAS Institute 1999). A forward stepwise multiple-regression model \( n = 285 \) sampling points for ACAD, \( n = 378 \) sampling points for GRSM) was used with forested deposition scaling factor as the dependent variable and several independent variables: elevation, \( (\text{elevation})^2 \), slope, and topographic-exposure dependent variable and several independent variables: used with forested deposition scaling factor as the multiple-regression model (analyses were conducted using the SAS statistical

general linear model (GLM) was then constructed with the landscape variables obtained from the stepwise regression: tree diameter, tree height, tree-canopy openness, and distance from coast variables, as well as dummy variables for categorical vegetation and aspect class variables. In ACAD, additional independent variables were available for the stepwise regression: tree diameter, tree height, tree-canopy openness, and distance from coast variables, as well as dummy variables for the presence of krummholz vegetation, or occurrence of fire in 1947. A general linear model (GLM) was then constructed with forested deposition scaling factor as a function of the landscape variables obtained from the stepwise regression, as well as significant interaction terms that increased model \( r^2 \). Deposition scaling factors were regressed against elevation for the five ACAD nonforested locations.

GIS-statistical model and map

Our second goal was to create an empirically based, combination GIS-statistical model in which the independent variables were restricted to only those variables currently available as GIS data layers. We then used this GIS-statistical model to create a new GIS data layer of deposition (LandMod, Fig. 1).

After removing independent variables (IVs) not currently available as GIS data layers, such as tree diameter, canopy openness, and presence of krummholz, GLM parameter estimates were recalculated. Using map algebra within GIS software, the GLM parameter estimates from the above statistical analyses were applied to each forested pixel (having coordinates \( x \) and \( y \) of the GIS database to calculate each pixel’s deposition SF (Fig. 1):

\[
\text{modeled SF at } x, y = [b_0 (at \ x, y)] + [b_1 \times (Z_1 \ at \ x, y)] + [b_2 \times (Z_2 \ at \ x, y)] + [b_3 \times (Z_1 \ at \ x, y) \times (Z_2 \ at \ x, y)] + \cdots + [b_n \times (Z_n \ at \ x, y)]
\]

where \( Z_1, Z_2, \ldots, Z_n \) are independent variables, \( b_0 \) is the intercept, \( b_1 \) is the landscape variable \( Z_1 \) coefficient, \( b_2 \) is the landscape variable \( Z_2 \) coefficient, and \( b_3 \) is the \( Z_1 \times Z_2 \) interaction coefficient.

Scaling factors for nonforested pixels in ACAD were calculated using the same procedure, but with different coefficients and landscape variable(s). Scaling factors for nonforested pixels in GRSM were assigned “no data” values.

This map of predicted scaling factors was then multiplied by reference deposition (RD) to produce map(s) of estimated annual deposition:

\[
\text{modeled deposition (time } T, \text{ ion } I \text{ at } x, y \text{) at } x, y = \text{ RD (time } T, \text{ ion } I \text{) } \times \text{ predicted SF at } x, y. \quad (2)
\]

For the vegetation type, slope, and aspect variables, where both field- and GIS-derived data were available, the field-derived data were deemed more accurate than GIS data and were therefore used in developing the GIS-statistical model, but by necessity the GIS versions of these variables were used to generate the deposition maps. For instance, if the GIS vegetation data layer showed a location to be under coniferous canopy, but our field data showed that the location was under deciduous canopy, we used the deciduous classification in creating the GIS-statistical model.

Map validation

Sulfur deposition residuals were plotted to examine the behavior of the LandMod model. At ACAD, a random subset of 30 sample locations was withheld from the full ACAD dataset and sulfur deposition residuals (measured throughfall subtracted from LandMod predicted) were plotted for those locations. At GRSM, throughfall collectors (109-day throughfall sampling period from 13 June 2000 to 30 September 2000) were colocalized with 32 forest-floor Pb sampling locations and the sulfur-deposition residuals (measured throughfall subtracted from LandMod predicted using forest-floor Pb index) were plotted.

Results

Deposition indices

Measured throughfall S deposition at Acadia National Park (ACAD) ranged from 0.85 to 9.83 kg S/ha for...
the period 7 June to 18 September 2000, with a mean of 2.54 kg S/ha. Measured deposition indices at the five nonforested ACAD locations ranged from 1.05 to 1.56 kg S/ha (Fig. 2a). Measured Pb concentration in the forest floor at Great Smoky Mountains National Park (GRSM) forested locations ranged from 7.06 to 122.8 mg Pb/kg, with a mean of 33.7 mg/kg (Fig. 2b).

**Fig. 2.** Spatial distribution of measured deposition indices (DI) for (a) sulfur flux in Acadia National Park (ACAD) (white boundary areas indicate seawater) and (b) forest-floor lead concentrations in Great Smoky Mountains National Park (GRSM). Larger bubbles indicate a higher deposition index; scale range categories overlap to reflect the impossibility of discerning fine differences in this type of map.

### Base deposition

Calculated base deposition at the ACAD monitoring station site for the period 6 June 2000 to 19 September 2000 was 1.14 kg S/ha (0.99 kg/ha of wet S + 0.15 kg/ha of dry S). The base deposition at the forest adjacent to the Great Smoky Mountains NADP site was 16.61 mg Pb/kg.
Deposition scaling factors

The scaling factors (SFs) for ACAD ranged 12-fold, from 0.76 to 8.76. The GRSM scaling factors ranged 17-fold: 0.43–7.40. Thus, hotspots of deposition are characterized by 12-fold higher deposition than “cold” spots of deposition at ACAD and 17-fold higher at GRSM. The spatial distribution of deposition SFs for the two parks indicates that many—but not all—of the high-elevation locations have high SF values (Fig. 2a, b).

Statistical Model Results

Deposition as a function of individual landscape independent variables.—The SFs were positively correlated with elevation at both ACAD ($r^2 = 0.21$, $P < 0.0001$, first-order fit, Fig. 3b) and GRSM ($r^2 = 0.42$, $P < 0.0001$, second-order fit, Fig. 4b). Scaling factors were also positively correlated with increased topographic exposure at both ACAD ($r^2 = 0.12$, $P < 0.0001$) and GRSM ($r^2 = 0.14$, $P < 0.0001$), although topographic exposure significantly covaried with elevation ($r^2 = 0.33$ and 0.30 at ACAD and GRSM, respectively). At GRSM, scaling factors increased with proximity to nearest road ($r^2 = 0.04$, $P < 0.0001$), but this relationship was not significant for the subset of locations less than 1 km from roads ($r^2 < 0.005$, $P < 0.63$). In contrast, at ACAD, scaling factors decreased with proximity to nearest road ($r^2 = 0.03$, $P < 0.01$).

Mean deposition enhancement, as indicated by mean scaling factors, at coniferous forest locations was higher than at deciduous locations at both ACAD ($P < 0.0001$) (Fig. 3a) and GRSM ($P < 0.0001$) (Fig. 4a). Scaling factors were not correlated with slope at either ACAD or GRSM. Scaling factors were also not correlated with aspect classes (centered on four cardinal directions) at ACAD but they were weakly correlated with the same aspect classes at GRSM; north aspect had significantly higher SFs than either west or east aspect in Tukey-adjusted multiple pairwise comparisons. At ACAD, SFs increased with increased qualitative canopy cover ($P = 0.004$), but at GRSM the SFs decreased with increased qualitative canopy cover ($P < 0.0001$). GRSM qualitative canopy-cover data were missing for 58 locations.

At ACAD only, the additional variables of distance to coast, tree species, tree height, tree diameter at breast height (dbh), presence of krummholz, quantitative canopy openness, and understory class were evaluated.
Actual S deposition decreased from as much as 13.2 kg S·ha⁻¹·yr⁻¹ (SF of 3.3) at 90 m from the coast to <6 kg S·ha⁻¹·yr⁻¹ of S at ~300 m from the coast, based on a transect of four collectors that extended from coast to inland, demonstrating a classic edge effect (e.g., Weathers et al. 1995, 2001). At the scale of the whole-park landscape, when all 285 data points were considered, there was a positive slope, but no statistically significant relationship between deposition and distance to the coast. There were significant differences in SFs under different tree species (P < 0.0001) as well: Picea spp. had the highest mean SF (2.9) and Acer pensylvanicum L. had the lowest mean SF (1.3, data not shown). Deciduous species had mean scaling factors <2 and coniferous species all had mean scaling factors >2, with the exception of Thuja occidentalis L. and Tsuga canadensis (L.) Carr., which had mean SFs of 1.7 and 1.5, respectively. Scaling factors had a positive correlation with tree height (P = 0.04), no correlation with tree dbh or presence of krummholz, and a weak negative correlation with quantitative hemispherical-photo canopy openness (P = 0.01). Scaling factors for locations with an understory were lower than for locations without an understory (P = 0.005), and for locations with an understory there were no significant differences in SF between coniferous and deciduous understories (P = 0.25).

At GRSM only, location with respect to the central topographic divide, as well as rhododendron understory presence, were evaluated as potential drivers of deposition. Deposition was somewhat higher on the Tennessee side of the central divide than on the North Carolina side (P < 0.01). Deposition decreased at GRSM with the presence of rhododendron, an evergreen, broad-leaved, understory plant (P < 0.0001; note: rhododendron data were missing for 53 locations), but the presence of rhododendrons is also uncommon at elevations >1800 m.

**Deposition as a function of multiple, landscape independent variables: StatMod.**—Stepwise multiple regression and general linear model (GLM) procedures for ACAD forests showed that elevation and forest type (coniferous or deciduous class) together explained 32% of the variation in deposition (P < 0.0001, Fig. 5a). When elevation, forest type, and elevation x forest type interaction terms were combined with tree dbh, the statistical model explained a total of 37% of the variation in deposition scaling factors (P < 0.0001, Table 2a). For the five nonforested open locations at ACAD there was a weak positive correlation of deposition SF with elevation in meters (deposition = 1.00760 + 0.00081(elevation), r² = 0.60, P < 0.12).

Stepwise multiple regression and GLM procedures for GRSM forests showed that elevation, (elevation)², and forest type (coniferous or deciduous class) together explained 46% of the variation in deposition (Fig. 5b). When these variables were combined with a forest type x elevation interaction term and slope (DEM-derived), the statistical model explained a total of 48% of the variation in our deposition indices (P < 0.0001, Table 2c).

**GIS-statistical model and scaling-factor maps.**—After randomly withdrawing 30 locations at ACAD for map validation purposes (see Discussion: How good are the maps?, below), stepwise regression and GLM procedures were used to select among the variables that were only represented in GIS data layers so that they could be used to generate deposition maps (Fig. 1, Table 2b). Of the variables selected for the statistical model in the previous step, at ACAD, krummholz presence and tree dbh did not have comparable GIS data, and this, combined with lower sample size (n = 255 data points), reduced slightly the explanatory power of the GIS-statistical relationships (r² = 0.31, P < 0.0001) for the GIS-statistical model.

When the scaling-factor parameter estimates from Table 2b were combined with the appropriate GIS data layers, the resulting deposition SF map for ACAD forests had values ranging from 1.1 to 4.5, a fourfold difference. Nonforested SFs based on the elevation regression ranged from 1.0 to 1.4.

At GRSM, 32 colocated deposition index locations were withdrawn for map validation purposes (Table 2d). The explanatory power of the resultant equations was
somewhat increased with the lower sample size ($n = 348$) ($r^2 = 0.51$, $P < 0.0001$). When the SF parameter estimates from Table 2d were combined with the appropriate GIS data layers, the resulting deposition SF map for GRSM had values ranging from 0.72 to 4.56, a sixfold variation.

**Modeled S and N deposition maps: LandMod.**—The ACAD scaling-factor map was multiplied by ACAD reference deposition of 3.00 kg N ha$^{-1}$ yr$^{-1}$ (2.51 kg N ha$^{-1}$ yr$^{-1}$ of wet deposition from NADP + 0.49 kg N ha$^{-1}$ yr$^{-1}$ of dry deposition from CASTNET) for all of calendar year 2000 to produce a 2000 modeled ACAD N-deposition map (Fig. 6). Nitrogen-deposition values ranged from 3.0 to 14 kg N ha$^{-1}$ yr$^{-1}$, with an area-weighted mean of 5 kg N ha$^{-1}$ yr$^{-1}$ of N for the entire ACAD study area (Fig. 6). For sulfur, the ACAD scaling-factor map was multiplied by ACAD reference deposition of 5.55 kg S ha$^{-1}$ yr$^{-1}$ (4.87 kg S ha$^{-1}$ yr$^{-1}$ of wet deposition + 0.68 kg S ha$^{-1}$ yr$^{-1}$ dry deposition) for all of calendar year 2000 to produce a 2000 modeled ACAD S-deposition map (Fig. 6). Sulfur-deposition values ranged from 5.6 to 25 kg S ha$^{-1}$ yr$^{-1}$, with an area-weighted mean of 9.5 kg S ha$^{-1}$ yr$^{-1}$.

Using the same method as for ACAD, the GRSM scaling-factor map was multiplied by GRSM reference deposition of 6.8 kg N ha$^{-1}$ yr$^{-1}$ (2.9 kg N ha$^{-1}$ yr$^{-1}$ of wet deposition + 3.9 kg N ha$^{-1}$ yr$^{-1}$ of dry deposition from the NADP and CASTNET programs, respectively) for calendar year 2000 to produce a 2000 modeled GRSM N-deposition map (Fig. 7). Nitrogen-deposition values ranged from 5 to 31 kg N ha$^{-1}$ yr$^{-1}$, with an area-weighted mean of 10 kg N ha$^{-1}$ yr$^{-1}$ for the entire park area. For sulfur, the GRSM scaling-factor map was multiplied by GRSM reference deposition of 9.1 kg S ha$^{-1}$ yr$^{-1}$ of S (5.8 kg S ha$^{-1}$ yr$^{-1}$ of wet deposition + 3.3 kg S ha$^{-1}$ yr$^{-1}$ of dry deposition) for all of calendar year 2000 to produce a 2000 modeled GRSM S-deposition map. The resulting LandMod GRSM S-deposition map shows values that ranged from 7 to 42 kg S ha$^{-1}$ yr$^{-1}$, with an area-weighted mean of 14 kg S ha$^{-1}$ yr$^{-1}$ (Fig. 7).

**Validation of LandMod deposition maps**

Sulfur-deposition residuals (LandMod predicted minus throughfall measured) were plotted as a function of elevation to examine model function (Fig. 8a and b). Thirteen out of 30 validation locations (43%) at ACAD and 13 out of 32 validation locations (41%) at GRSM had mismatches in vegetation type between GIS and field data and were converted to the field value before calculating residuals. After making those changes, the correlation between measured and predicted deposition

### Statistical model parameters of deposition scaling factor (unitless values that show relative deposition; see Fig. 1 and Methods) as a function of field- and GIS-measured (StatMod) landscape variables for (a) Acadia National Park, ACAD ($n = 285$ data points), and (c) Great Smoky Mountains National Park, GRSM ($n = 378$ data points). The table also reports mapping equation parameters of deposition scaling factor as a function of GIS-measurable landscape variables for (b) ACAD ($n = 255$ data points) and (d) GRSM ($n = 346$ data points).

<table>
<thead>
<tr>
<th>Variable</th>
<th>Coefficient</th>
<th>$P$</th>
<th>Partial $r^2$</th>
<th>Model $r^2$</th>
</tr>
</thead>
</table>
| **Acadia National Park, ACAD**
| a) Statistical model parameters | | | | |
| Intercept | 0.98720 | | | |
| Elevation (m) | 0.00265 | <0.0001 | 0.212 | 0.212 |
| Conifer presence | -0.03922 | 0.8676 | 0.105 | 0.317 |
| Tree dbh (cm) | 0.01521 | 0.0098 | 0.019 | 0.336 |
| Elevation × conifer pres. | 0.00482 | 0.0003 | 0.031 | 0.367 |
| b) Mapping equation parameters | | | | |
| Intercept | 1.60345 | | | |
| Elevation (m) | 0.00263 | 0.0193 | 0.171 | 0.171 |
| Conifer presence | -0.03248 | 0.9061 | 0.080 | 0.251 |
| Mixed forest presence | 0.48995 | 0.0380 | 0.018 | 0.269 |
| Distance to coast (m) | -0.00015 | 0.0399 | 0.006 | 0.275 |
| Elevation × conifer pres. | 0.00481 | 0.0008 | 0.032 | 0.307 |
| **Great Smoky Mountains National Park, GRSM**
| c) Statistical model parameters | | | | |
| Intercept | 3.32326 | | | |
| (Elevation)$^2$ | 0.000001995 | <0.0001 | 0.322 | 0.322 |
| Elevation (m) | -0.00427 | <0.0001 | 0.096 | 0.418 |
| Conifer presence | -0.15380 | 0.6113 | 0.044 | 0.416 |
| Slope (from DEM) | 0.01106 | 0.0250 | 0.007 | 0.468 |
| Elevation × conifer pres. | 0.000648 | 0.0070 | 0.008 | 0.479 |
| d) Mapping equation parameters | | | | |
| Intercept | 3.69836 | | | |
| (Elevation)$^2$ | 0.000002195 | <0.0001 | 0.331 | 0.331 |
| Elevation (m) | -0.00464 | <0.0001 | 0.125 | 0.455 |
| Conifer presence | -0.26948 | 0.3737 | 0.044 | 0.499 |
| Elevation × conifer pres. | 0.000748 | 0.0030 | 0.014 | 0.513 |
was quite high for ACAD ($r^2 = 0.64$, $P < 0.0001$) and GRSM ($r^2 = 0.80$, $P < 0.0001$), although it was not a 1:1 relationship. At both ACAD and GRSM, LandMod slightly overestimated deposition at low elevations and underestimated deposition at high elevations.

The residual plots show interesting and consistent patterns between the two sites. The model behaves differently at low-elevation, low-deposition sites than at high-elevation, high-deposition sites, and for different vegetation types. At ACAD, the model overpredicts at low elevations, more so for coniferous and mixed vegetation than for deciduous. The residuals were closer to zero for deciduous vegetation at low elevations. At high elevation, the model underpredicts for coniferous vegetation. In the subset of data used for validation there is little representation of deciduous vegetation at high elevation. For GRSM, the model also overpredicts at low elevation for deciduous vegetation (the primary vegetation type at low elevation) and under predicts rather significantly for high-elevation conifer forests, relative to field measurements.

As a further validation of the ACAD model, we compared the LandMod predicted deposition with sulfur deposition from a frequently used regional model of deposition (ClimCalc [available online]; Ollinger et al. 1993), as well as with reference data from the NADP and CASTNET monitoring stations (Fig. 9). Although our validation data suggest that the LandMod underestimates high deposition at high elevations, this analysis demonstrates that the existing estimates for those locations are considerably lower than what our approach produces (Fig 9).

**DISCUSSION**

**Indices of deposition**

To estimate spatial patterns of atmospheric deposition, our surrogate, or index, measures of total deposition were sulfate in throughfall and lead in forest floor. In general, important criteria for the use of surrogates for total deposition are that the surrogate element or ion used (1) is relatively biologically inert, (2) preserves the pattern of deposition, and (3) represents total (rain, snow, dry, cloud/fog) deposition.

Throughfall is the movement of water from the forest canopy to the forest floor. Throughfall S has been shown to be a robust surrogate measure of total S deposition (Lindberg and Lovett 1992). The strengths of throughfall (TF) are that it encompasses all forms of deposition (wet, gaseous, particulate, fog), and does not involve assumptions about canopy or terrain characteristics. The major weakness of TF in our application is that it should be sampled continuously over all seasons and for many years to reflect more accurately the long-term, average patterns of deposition. Throughfall measurements also can be labor intensive to collect, and stemflow (SF, water that moves from the canopy down the stems of plants [to the forest floor]) should be included (although often it is a small percentage of total flux) but is more difficult to sample. Furthermore, TF and SF could be influenced by leaching or uptake of S in the canopy, although this has been shown to be minor in most studies (Lindberg and Garten 1988, Cape et al. 1992).

In this study, because of the difficulty of accessing several hundred (~300) TF collectors in roadless areas, it was possible to measure throughfall S only during the summer growing season, when deciduous trees have leaves. In creating annual deposition maps from these data, it was necessary to assume that the landscape patterns of deposition for the entire year are similar to the patterns we measured during the leaf-on season. An analysis of the four-year average (1999–2003) NADP plus CASTNET deposition data by season and by element (S and N) showed surprisingly little variation in wet-to-dry ratios: N dry deposition varied 3% and 6% of total deposition across seasons, and S dry
deposition varied between 3% and 10%. Nonetheless, we recognize that seasonal variations in leaf area, prevailing wind direction (aspect effect), and fog frequency and extent can influence deposition patterns, so we recommend that future research be directed at measuring TF + SF fluxes year-round. We also note that winter conditions (i.e., deciduous leaf drop) can be readily simulated in LandMod to account for a known change in leaf area.

Lead in the forest floor has also been used to indicate regional patterns of pollution (Johnson et al. 1982, Graustein and Turekian 1989) as well as more-local deposition patterns (Weathers et al. 1995, 2000). Lead, or any other atmospherically deposited and biologically inert substance that is used to indicate the deposition of some other element (S, for example) should be tested for its appropriateness. This is especially true for the pollutant Pb, whose emissions have been reduced significantly since the 1970s (Johnson et al. 1995). Once Pb has been deposited to the forest floor it moves, though usually quite slowly (decades to centuries), from forest floor to mineral soil (e.g., Wang et al. 1995). Recent reports have documented faster-than-expected Pb movement from forest floor to mineral soil (Johnson et al. 1995, Kaste et al. 2003, Watmough et al. 2004), which suggests the need for some independent measure of the relationship between this deposition index and atmospheric deposition to support its use as a deposition index. Although we previously demonstrated the utility of Pb as an index of deposition (Lovett 1994, Weathers et al. 1995, 2000), for this work we performed an additional test by measuring Pb concentrations in forest floor samples from forested sites adjacent to seven NADP monitoring stations. As noted in Methods (see Field methods for Great Smoky Mountains National Park, above), these locations span a 3–4 fold depositional gradient, as well as a large latitudinal gradient. The concentration of Pb in forest floor at these sites was strongly correlated with total deposition of both S and N (P < 0.001), supporting its use for detecting patterns of deposition in this study.

One of the most significant values of Pb as a surrogate measure of atmospheric deposition is that continuous sampling is not necessary—Pb in the forest floor reflects long-term patterns of deposition, integrating both seasonal and interannual variations. It is possible to collect many more samples over a much larger area if samples need to be collected only once. The weakness is that, although Pb is carried on submicron particles that exhibit some characteristics of gas behavior, there are no true gaseous forms of Pb; therefore, it may not be an accurate index of pattern for substances in which the pattern of gaseous deposition is significantly different from that of particulate and cloud-water deposition. Finally, Pb in forest floor is truly a surrogate measure of deposition; here we were not interested in Pb deposition, per se, but rather, our goal was to use Pb as an indicator of patterns of S and N deposition.

Because it integrates deposition over time, and because sampling is logistically more feasible, we had intended to use only Pb as the index of deposition for this study at both parks. However, because approximately one third of the area of ACAD burned in the 1940s, the pattern of Pb in the forest floor may have been altered by fire. Therefore, we measured sulfate in throughfall as an index of deposition at ACAD.
Elevation and vegetation as landscape-independent variables

For both ACAD and GRSM, elevation and vegetation were the independent variables that explained most of the variance in the statistical models (StatMod) of deposition.

Elevation.—It is not surprising that elevation was found to be a primary control on the landscape pattern of deposition. Orographic effects drive precipitation increases with elevation (Lovett and Kinsman 1990). Dry deposition may also increase with elevation, because the total exposed surface area for collection of particles and gases is often greater (Stachurski and Zimka 2000) and windspeeds are higher, both of which can influence dry deposition (Lovett and Kinsman 1990). For inland mountain ranges, cloud-water deposition is greatest at high elevations (Lovett and Kinsman 1990, Weathers et al. 2000). Previous studies have shown nonlinear increases with elevation, and have attributed these step increases at mid-to-high elevation to cloud deposition, which occurs only above a clearly defined cloud base (Siccama 1974, Lovett et al. 1999, Weathers et al. 2000). As a result of increased wet, dry, and cloud-water deposition we had anticipated that total ion deposition would increase with elevation and that elevation would therefore be an important independent variable in our model. Since the elevation range at ACAD was <500 m, it is not surprising that the deposition relationship is weaker than at GRSM, where there was a 1500-m elevation range. Fog-deposition relationships with elevation are likely to vary between inland and coastal regions as well. This may further explain the stronger relationship of deposition with elevation at GRSM compared to ACAD. Casual observations suggest that fog deposition at ACAD near the coast may alternate between being highest at mountaintops and at low elevations, the latter when advective fogs move from ocean to land. Systematic data are needed to either refute or substantiate this observation, but if in fact there is only an intermittent cloud base of irregular elevation this could be a contributing factor that helps explain why ACAD had only a first-order elevation relationship with deposition while GRSM had a second-order relationship between elevation and deposition.

Vegetation.—It is also not surprising that vegetation was found to be a primary control of the landscape pattern of deposition. In general, evergreen coniferous vegetation and mixed coniferous–deciduous forests, with their greater leaf-area index and year-round collection surface, have much more efficient scavenging surfaces for particles (including cloud droplets) and gases than does most deciduous vegetation. A few studies have demonstrated differences in total deposition among these broad vegetation classes (Ivens 1990, Weathers et al. 1992, 1995, Lovett et al. 1999, Weathers et al. 2000). It is important to reiterate that the GRSM Pb index incorporates both the high magnitude and long duration of leaf-area collection surface for coniferous forests,

FIG. 8. Residuals of predicted vs. observed sulfur deposition plotted as a function of elevation for (a) Acadia (ACAD) and (b) Great Smoky Mountains (GRSM) National Parks. GIS and field vegetation data were matched for this analysis. Deciduous, coniferous, and mixed (ACAD only) stands are shown.

FIG. 9. Comparison of S deposition (103-day sampling period) at 32 validation data points from Acadia National Park (ACAD; see Results for description of validation points) using the LandMod described in this paper, deposition estimates for each of the points based on Ollinger et al. (1993), and the NADP and CASTNET programs; these monitoring data have only one value for all locations in the region. The 1:1 line is also shown.
while we are using the flux of S in summer throughfall S to model annual deposition at ACAD.

At ACAD, we were able to discern differences in deposition among species as indicated by the relative difference in scaling factors. These data confirm that most coniferous canopies are better scavengers than deciduous canopies—there is on the order of twofold enhancement.

Attempts to correlate leaf area with deposition produced inconclusive, and in some cases unexpected, results. Qualitative identification of ACAD leaf area as low, intermediate, or high was a slightly better predictor of deposition than quantitative canopy openness from hemispherical photos, presumably because the latter did not take into account overlapping leaf-area surfaces in the upper canopy. Both measures suggested that increased leaf area was associated with increased deposition; these correlations were weak but in the expected, positive direction. By contrast, a qualitative assessment of canopy cover at GRSM suggested that increased leaf area was (weakly) associated with decreased deposition. In addition, locations with an understory had slightly lower deposition at both ACAD and GRSM. These results are not readily explicable with the data that we have; more appropriate quantitative measures of vegetation structure relevant to deposition of dry and fog deposition are needed.

**Elevation–Vegetation interaction.**—One of the benefits of this study is that by sampling hundreds of locations it was possible to simultaneously address elevation and vegetation variables. The strong interaction between vegetation type and elevation we observed reflects the much stronger relationship between elevation and deposition for coniferous and mixed forests compared to deciduous forest. Higher wind speeds at high elevation create the potential for higher dry deposition. Increased clouds at high elevation (for locations with a distinct cloud base) create the potential for higher fog/cloud deposition. It could be argued that this potential for high deposition at high elevation is fully captured only where there are coniferous forests with high leaf area (Weathers et al. 2000).

**How good are the maps?**—There are several important factors that determine the accuracy of the deposition maps produced here. Errors in sampling and analyzing point measurements of deposition chemistry would decrease accuracy, but results from our quality-control procedures seem to be reasonable, and the validation analyses when the erroneous GIS data were corrected to match the observed field data. It seems reasonable to assume that, as remotely sensed data with a resolution of 1 m or better become available, the number of mismatches between GIS and field vegetation should decrease. The sources of error described above must be taken into consideration. However, we think that the greatest and most interesting sources of uncertainty in the deposition maps arise from as-yet unquantified or unidentified variables that cause real variability in deposition—variables that are related but not part of our StatMod or LandMod, but are evident in the
pattern displayed in our validation data (Fig. 8a, b, NIST/SEMATECH 2005).

The statistical relationships between independent variables and deposition indices were highly significant, yet they explained less than half of the variance in the data. What explains the rest of the variance in actual deposition? It is likely that the independent variables that explain the rest of the variance, at a broad scale, are those that affect the capture of particles and gases by vegetation, such as canopy roughness and orientation and topographic exposure. Everything else held equal, there is an optimum canopy leaf area and morphology for maximum deposition. For example, Lovett and Reiners (1989) modeled cloud deposition as a function of canopy leaf-area index (LAI) and found highest deposition at a mid-range LAI. Our data suggest a similar enhancement for predominately coniferous forest—those regions with up to 25% deciduous canopies—compared with pure deciduous or coniferous forest at ACAD (Fig. 3a); however, this was not true at GRSM (Fig. 4a). Canopy and leaf architecture and morphology are also likely to influence deposition. Our limited data from ACAD showing large differences in fluxes among coniferous trees suggests that there may be factors in addition to LAI that control deposition. To wit, the highest measured deposition in the ACAD validation data set—the biggest outlier for predicted vs. measured values—was a large-statured (7-m-tall) conifer that was situated on an exposed ridge. This is just the kind of tree-specific—or site-specific—depositional environment that no general model can readily replicate.

A major variable controlling small-scale patterns in deposition is likely to be vegetation exposure, including trees whose crowns are exposed above their neighbors and trees growing in unsheltered locations such as ledges and promontories. A tree with high exposure is likely to get much higher deposition than another tree of equivalent LAI and height that is sheltered by neighbors. This has been demonstrated in several studies of forest edges (Weathers et al. 1992, 1995, 2001, Lindberg and Owens 1993). While we were able to generate a GIS index of topographic exposure using elevation models of the ground surface, we believe the horizontal and spatial resolution of those data are not yet good enough to generate an index of the combination of canopy architecture and exposure that is relevant to rates of deposition. However, the development of high-resolution remotely sensed LIDAR (light detection and ranging) data shows promise in indicating both the elevation of tree canopies and the density of tree canopies, which should help considerably in filling this data gap. Another challenge is the scale at which topographic exposure might matter, which is likely to be less than 10s of meters rather than the 30 × 30-m pixel resolution of most GIS data layers. If it were possible to measure canopy architecture and exposure (or surrogates) easily, and better yet, if they were available as GIS data layers, landscape models of deposition could probably be made more accurate. Thus an important next step for deposition research will be to develop a way to quantify different canopy LAIs and architectures along with their exposure, perhaps using remotely sensed data (e.g., LIDAR), and examine their relationship to deposition.

**Map validation**

The residual data show that at ACAD, the LandMod map most significantly underestimated deposition in high-elevation regions; these points also represent the highest measured deposition values. The three data points that represent high-elevation conifer forests show the biggest discrepancies. The GRSM validation plots show much bigger discrepancies for high-elevation regions, indicating that high-elevation data reduce the homogeneity of variance for the LandMod model. That the variance is high, especially at high elevation, is unsurprising given the large scatter of the primary data (Fig. 3b).

We cannot say definitively how much of an influence our use of Pb in forest floor to create the LandMod had on the validation comparison. However the consistency between the two validation plots (Fig. 8a, b) suggests that there are missing variables in LandMod rather than an effect of using different deposition indices at ACAD vs. GRSM. We might predict that LandMod would underestimate tree-with-leaf-on patterns of S deposition (i.e., the measured values) since the Pb index integrates deposition over seasons and years. It is likely that actual sulfur deposition for a short time during the season of highest deposition (i.e., when we made the S flux measurements for the validation data) would be higher than predicted deposition. In addition, LandMod estimates deposition to a 30 × 30 m pixel that is characterized by an average elevation and vegetation type. We measured deposition to a small area that surrounded individual trees.

Clearly these models need refinement to be able to accurately predict deposition to heterogeneous landscapes. The validation data sets can be used to suggest next steps, such as the need to identify and quantify controls on deposition to high-elevation conifer forests. However, it is important to consider our results within the perspective of the data that are currently available. LandMod appears to be conservative: it slightly over-predicts for low-elevation, low-deposition environments, but significantly underestimates input for high-elevation, high-deposition environments. The deposition data that are currently available for these landscapes—exclusive of our LandMod—either show no spatial variability (for example, GRSM has one NADP + CASTNET value for the whole park), or some spatial variability over the region (i.e., the Ollinger et al. [1993] regional-deposition model for the northeastern United States), but do not capture spatial variability at the scale of one to hundreds of kilometers. Thus, LandMod S deposition for ACAD, compared with S predictions based on Ollinger et al.
(1993; a widely used regional deposition model (see also footnote 8), and with the monitoring-station data alone, captures the response of measured deposition to both elevation and vegetation type that the other two estimates cannot (Fig. 9). For GRSM, in addition to the monitoring station wet + dry deposition estimate, cloud deposition was estimated at the peak of Clingman’s Dome for the time period over which we sampled (G. M. Lovett, unpublished manuscript). For this high-elevation region, the predicted cloud + wet + dry deposition based on the monitoring-station data, 27 kg S/ha for a four-month growing season, is more than twice the S measured in the throughfall at Clingman’s Dome. Our LandMod predicts less than half the measured S in throughfall for this same location. Again, it appears that our measured as well as our modeled deposition is quite conservative (i.e., underestimates total deposition) compared to existing data.

How transferable is this approach?

The empirical scaling factors and models we have created are likely to be applicable to other chemical species that are deposited from the atmosphere in approximately the same proportions and by the same mechanisms as N and S are deposited. In addition, where strong and predictable relationships can be established between chemical species \( x \) and \( S \), the models might be extended to other atmospherically deposited nutrients and pollutants.

Our approach may also be applicable to other sites. We think that elevation and vegetation type are likely to be important driving variables for deposition in other mountain ranges around the world, albeit the coefficients may differ from landscape to landscape depending upon such factors as the dominant mode of deposition, for example fog deposition, dry deposition, and whether snow is a dominant proportion of wet deposition, patchiness of the landscape, and its elevational span. Both of these topics—application of the model to other chemical species and to other sites—warrant further research and comparison.

Updating maps

The statistical relationships that underpin the atmospheric-deposition maps developed here are not expected to change from year to year; the independent variables that drive LandMod (vegetation and elevation) are likely to be stable across the landscape. However, major vegetation changes as a result of an anthropogenic disturbance such as logging, or natural disturbances such as fire, major storms or insect outbreaks would affect the spatial pattern of deposition. These large-scale changes could be accommodated by recalculating scaling factors. On the other hand, reference deposition data from the NADP and CASTNET monitoring stations do change over time. Since reference deposition data are incorporated in the last step of creating the deposition map, it is straightforward to update the deposition maps to reflect this temporal change. If data layers with finer resolution or other key data become available, they too could be substituted for the old data layers at the stage where scaling-factor maps are generated (Fig. 1).

Who cares?

Our park-wide LandMod deposition maps are based on spatially explicit inputs, and as such they can be used to identify regions of high and of low deposition (hotspots and coldspots). But, at the scale of the whole park, does it matter? Do we need to have spatially explicit estimates of deposition? One way to answer this question is to compare our area-weighted total deposition with currently available data from the NADP and CASTNET monitoring stations. Our models indicate that total, area-weighted N deposition is much greater than suggested by the local monitoring-station data (e.g., Fig. 9). ACAD has 70% greater, and GRSM 50% greater, N deposition when our LandMod is compared to NADP + CASTNET deposition estimates. These results demonstrate that even the average total deposition across mountainous landscapes is substantially underestimated by using data from single-point, low-elevation monitoring stations.

At places such as GRSM where a complete, spatially explicit biologic inventory is being compiled, it is possible to overlay deposition maps of hotspots with known sensitive species distributions to identify populations that are likely to be at greatest risk of damage from air pollution. Knowledge of deposition patterns can also be used to good advantage in designing effects research. For instance, identifying strong deposition gradients in areas where other factors (e.g., soil type or temperature) are relatively constant could facilitate comparative studies of the effects of deposition on plants and animals. In general, knowing spatial variability in deposition across a landscape can enhance tests of ecological responses to a range of inputs.

Conclusion

Estimates of atmospheric deposition in complex terrain are currently inadequate; they are either incomplete (spatially representative, but only for wet deposition, for example), or not spatially explicit. In this research, we have developed an empirically based model for scaling up to the landscape (tens to hundreds of kilometers) from point measurements to get spatially explicit estimates of atmospheric deposition. Our LandMod was created by applying a statistical model to GIS data layers. National monitoring network data were used for reference deposition; the deposition estimate from which to scale up. The resultant maps show several-fold variation in deposition across the landscape of two national parks. Vegetation type and elevation are the independent variables that most strongly controlled deposition.
This modeling effort and our validation exercises and explorations were revealing: they confirmed that there is large variance in deposition, especially at high elevations and that, with current tools, landscape-scale (tens to hundreds of kilometers) models are unlikely to be able to capture extreme deposition over areas of small spatial extent. Future research should test LandMod in other mountainous environments, expanding it to other chemical species, and refining it to account for (currently) unexplained variation in deposition. We have offered suggestions for how this might be accomplished. We predict that the latter effort will benefit significantly as new remote sensing technology and resulting GIS data layers become available and are field tested.

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LITERATURE CITED


