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Estimates of critical acid loads and exceedances for forest soils across the conterminous United States

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This simple mass balance equation estimated that 17% of US forest soils exceed their critical acid load by more than 250 eq ha⁻¹ yr⁻¹, and these areas are predominantly located in the northeastern US.

Abstract

Concern regarding the impacts of continued nitrogen and sulfur deposition on ecosystem health has prompted the development of critical acid load assessments for forest soils. A critical acid load is a quantitative estimate of exposure to one or more pollutants at or above which harmful acidification-related effects on sensitive elements of the environment occur. A pollutant load in excess of a critical acid load is termed exceedance. This study combined a simple mass balance equation with national-scale databases to estimate critical acid load and exceedance for forest soils at a 1-km² spatial resolution across the conterminous US. This study estimated that about 15% of US forest soils are in exceedance of their critical acid load by more than 250 eq ha⁻¹ yr⁻¹, including much of New England and West Virginia. Very few areas of exceedance were predicted in the western US.

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1. Introduction

The potential impacts of acid deposition on forests caused much concern among environmental scientists and policy makers across New England in the mid-1980s and early 1990s. Reports of high-elevation forest mortality prompted numerous studies to assess the short- and long-term effects of

nitrogen (N) and sulfur (S) compounds (the two principle components of acid deposition) on ecosystem health. Research studies suggested that long-term deposition of acidic compounds could negatively impact stream, lake, forest sustainability, and biodiversity (Wright and Schindler, 1995).

The Clean Air Act Amendments (CAAA) were enacted to address acidic deposition impacts on human and ecosystem health (CAAA, 1990). The CAAA primary focus was to reduce industrial emissions of S but mandated little change in N emissions (CAAA, 1990). Since the passage of the CAAA, S deposition has decreased by as much as 30% across the eastern US, but N deposition has changed little from 1980s' levels (Baumgardner et al., 2002).

During the 1980s, European forests were also receiving high rates of acidic deposition (Cowling, 1981), and European nations began using simple mass balance equations (SMBEs)

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to estimate forest soil critical acid loads (CALs) (Hall et al., 2001; Posch et al., 2001). A CAL can be defined as a quantitative estimate of ecosystem exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur, according to present knowledge (Nilsson and Grennfelt, 1988; Gregor et al., 2004). When pollutant loads exceed the CAL there is an increased risk of harmful ecosystem impacts. Pollutant load (in the form of acidic deposition) in excess of the CAL is termed the exceedance (Gregor et al., 2004). As exceedance increases, the potential for negative forest health impacts also increases (Sverdrup and De Vries, 1994). The objectives of European and, more recently, Canadian studies were to circumscribe areas where N and S loads were potentially degrading forest health.

Research studies suggest that ecosystems with high rates of acidic deposition and low CAL are more likely to experience nitrate leaching, forest mortality, reduced forest productivity, reduced biological diversity, and increased stream acidity (Driscoll et al., 2001). Whereas forest soil CAL and exceedance studies have been developed on national-scales for both Europe and Canada, much less research has been conducted on CAL for forest soils in the United States (US) (Aherne and Watmough, 2005). The objective of this study was to use an SMBE to estimate the current CAL and exceedance of the CAL for forest soils across the conterminous US. The intention was to locate forest soil areas that could potentially be in exceedance of the CAL as a first step toward an assessment of the long-term impacts of acidic deposition on forest ecosystem health in the conterminous US.

2. Methods

2.1. Simple mass balance equation

National-scale vegetation, soil, and climate databases were compiled to estimate the critical acid load for forest soils. CAL was modeled using a steady state, SMBE of ecosystem N and S inputs, sinks, and outputs. An SMBE assumes that the ecosystem is at equilibrium over time, and that the point of equilibrium is equal to the ecosystem's CAL. The estimates of forest soil CAL and exceedance were modeled within a geographic information system (GIS), where each SMBE parameter was represented by data on a 1-km² grid system. The forest soil CAL of N and S was expressed in equivalents per hectare per year (eq ha⁻¹ yr⁻¹) and calculated using the following equation:

$$\text{CAL}(S+N) = \text{BC}_{\text{dep}} - \text{Cl}_{\text{dep}} + \text{BC}_{\text{w}} - \text{BC}_{\text{u}} + \text{N}_i + \text{N}_u + \text{N}_{\text{de}} - \text{ANC}_{\text{le.crit}} \quad (1)$$

where CAL(S+N) is the forest soil critical acid load for S and N; BC_{dep} is base cation (i.e., calcium (Ca) + potassium (K) + magnesium (Mg) + sodium (Na)) deposition; Cl_{dep} is chloride deposition; BC_w is base cation weathering; BC_u is uptake of base cations (i.e., Ca + K + Mg) in trees; N_i is nitrogen immobilization; N_u is uptake of nitrogen in trees; N_{de} is denitrification; and ANC_{le.crit} is forest soil acid neutralizing capacity of CAL leaching (Gregor et al., 2004).

CAL exceedance (eq ha⁻¹ yr⁻¹) was calculated using the following equation:

$$\text{Ex}(S+N_{\text{dep}}) = S+N_{\text{dep}} - \text{CAL}(S+N) \quad (2)$$

where Ex is exceedance of the forest soil critical N and S loads; S + N_{dep} is the deposition of S + N; and CAL(S + N) is the forest soil critical load of S + N (Gregor et al., 2004). Higher Ex-values reflect greater exceedance of acidic deposition above the level associated with an increased likelihood of environmental harm.

3. Databases

Input data for the SMBE with the finest possible spatial resolution and regional to national extent were selected and compiled. All data sets except dry deposition were acquired in a GIS grid format. The dry deposition data were acquired in a point format and interpolated with ArcGIS using the nearest neighbor method (Minami, 2000).

The soil map unit database was used as the base grid, and all other spatial data were aligned to the soil map by synchronizing the grid's spatial extent. This ensured that each grid had the same number of rows and columns, and that the geographic location of each cell did not vary between the data sets. All data sets were originally mapped at different scales (Table 1) and resampled to 1-km² using the nearest neighbor method (Minami, 2000). This method was chosen because the cell value remains the same throughout the processing. However, the interpretation of the modeled outputs is limited by the coarsest spatial resolution of the original database.

3.1. Deposition

The National Atmospheric Deposition Program/National Trends Network (NADP/NTN) collects wet atmospheric deposition at 250 monitoring sites located across the US. NADP/NTN developed annual isopleth maps of ion concentration, precipitation, and total ion deposition for the continental US (NADP, 2005). The average ion deposition isopleth grids for the western US from 1994 through 2000 were input into the SMBE after being resampled from 2.5 km² to 1-km² using the nearest neighbor method.

Grimm and Lynch (2004) created a high-resolution wet deposition map of the eastern US that produced estimates with less error than 2D multiquadric algorithm or kriging model. Their model integrated National Oceanic and Atmospheric Administration (NOAA) precipitation data, precipitation

Table 1
Original resolution of input databases

Data set	Source	Original display scale	Original grain (cell size)
Dry deposition	CASTNET (US EPA, 2007)	n/a	n/a
Forest type	National Atlas (USFS and USGS, 2000)	1:7,500,000	1.1-km
Runoff	Gebert et al. (1987)	1:7,500,000	1-m
Soils	STATSGO (USDA NRCS, 1995)	1:250,000	6.25-km
Wet deposition	Grimm and Lynch (2004)	n/a	330-m or 1-km
	NADP annual isopleth maps (NADP, 2005)	n/a	2.5-km

chemistry data from NADP/NTN sites, and elevation, slope, and aspect data to estimate wet deposition. Wet deposition was estimated east of the 94°W longitude and from 25°N to 49°N latitude. The spatial resolution of the modeled data ranged from 353 m² at 25°N to 300 m² at 49°N (Grimm and Lynch, 2004). Both of these data sets were aggregated to a 1-km² resolution. The Grimm and Lynch data for 1994 to 2000 were input into the SMBE after averaging the years and combining the data set with the NADP/NTN isopleth data for the western US (NADP, 2005). Wet deposition was expressed in eq ha⁻¹ yr⁻¹ for S, N, base cations (i.e., Ca, Mg, and K), and Cl.

Gregor et al. (2004) suggested that wet deposition data used for estimating forest soil CAL should be corrected for sea-salt within 70 km of the coast. We applied the methods prescribed by Gregor et al. (2004) for this correction to the Atlantic, Gulf, and Pacific coastlines. However, the method clearly over corrected the wet deposition estimates (data not shown) in some areas. Therefore, we elected to use the uncorrected wet deposition data, acknowledging that the SMBE may have over estimated forest soil CAL within 70 km of the coast. Additional work is needed to develop equations that will accurately correct wet deposition for marine sources throughout all coastal areas of the US.

The Clean Air Status and Trends Network (CASTNET) has approximately 80 monitoring sites throughout the US that collect both dry and wet deposition data (US EPA, 2006). Sulfur and nitrogen dry deposition surfaces were interpolated to a 1-km² spatial resolution from the CASNET dry deposition point data using ordinary kriging. Cloud deposition data were not used in predicting CAL in this study due to the lack of availability of cloud data sets and the complexity in spatially interpolating these forms of deposition. The authors acknowledge the limitation of using only wet and dry deposition in calculating forest soil CAL but determined that the uncertainty associated with cloud deposition inputs would not improve the spatial accuracy of SMBE estimates of forest soil CAL exceedance areas. Methods for reducing spatial resolution errors associated with cloud deposition and spatial distribution are in progress, and cloud deposition may be incorporated into forest soil CAL estimates in the future.

3.2. Base cation weathering

The base cation weathering rate was estimated using the clay correlation-substrate method (Sverdrup et al., 1990). This method used a combination of parent material and clay percent to determine the weathering rate. Base cation weathering rate (eq ha⁻¹ yr⁻¹) was calculated as:

$$\text{Acid substrate: } W = (56.7 \times \% \text{clay}) - (0.32 \times \% \text{clay}^2)$$

$$\text{Intermediate substrate: } W = 500 + (53.6 \times \% \text{clay}) - (0.18 \times \% \text{clay}^2)$$

$$\text{Basic substrate: } W = 500 + (59.2 \times \% \text{clay}) \quad (3)$$

where W is base cation weathering. A temperature correction can be applied to this method, but this correction is more suitable for northern climates and was not used in the model.

The Earth System Science Center (ESSC) at Pennsylvania State University developed CONUS-SOIL, a 1-km² multi-layer soil data set based on the Natural Resource Conservation Service (NRCS) State Soil Geographic Data Base (STATSGO) (Miller and White, 1998). The STATSGO data set was a compilation of geology, topography, vegetation, climate, Landsat Thematic Mapper (TM) satellite imagery, and detailed, county-level soil survey data. Each soil map unit in STATSGO includes multiple components and data layers (USDA NRCS, 1995). STATSGO is organized by state and consists of one geospatial vector representing the soil map units for that state and 15 tables describing characteristics of those map units. Multiple soil layers are associated with each map unit. Soil layer sampling depth is not consistent within a state or between states. ESSC converted the vector map unit layer to a 1-km² grid, remapped many of the original STATSGO attribute layers, and defined 11 standard soil layers (Miller and White, 1998; Table 2). These data layers and tables linked the standardized data set to the original STATSGO data set distributed by ESSC as 1-km² soil map unit grids for the conterminous US. The CONUS-SOIL was much better suited for national-scale forest soil CAL modeling than the original STATSGO attribute layers and was therefore used in this study. Key soil data inputs for the CAL SMBE included CONUS-SOIL map units and clay fraction (Miller and White, 1998).

The forest soil percent organic matter (OM) layer was created using the ESSC technique for remapping STATSGO layers into CONUS-SOIL layers. First, the maximum and minimum recorded values for OM were averaged for each layer in the STATSGO data set. Second, the average OM layers were remapped into the 11 standard CONUS-SOIL layers using a weighted average to redistribute the average OM STATSGO layers into the CONUS-SOIL layers. If a STATSGO layer was completely contained in a CONUS-SOIL layer, then the average OM was multiplied by the component percent to determine the average OM contribution to the standard layer. If the STATSGO layer overlapped more than one CONUS-SOIL layer, then the proportion of overlap was multiplied by the average OM and the component percent, where the

Table 2
CONUS-SOIL standard layers (Miller and White, 1998)

Standard layer	Thickness (cm)	Depth to bottom of layer (cm)
1	5	5
2	5	10
3	10	20
4	10	30
5	10	40
6	20	60
7	20	80
8	20	100
9	50	150
10	50	200
11	50	250

component percent was the proportion of the soil map unit comprised of that soil component. Once the conversion from STATSGO to CONUS-SOIL layer was complete, the 11 standard layers were summed by layer and divided by the sum of component percent. Finally, the weighted average was calculated (Eq. (4)) to obtain one OM value per soil map unit.

Clay fraction was derived from a weighted average of soil fraction per map unit, using the following equation:

$$\text{Clay fraction of soil} = \sum s_{1-11} \times t / \text{depth to bedrock layer} \quad (4)$$

where s_1 through s_{11} is the percent of clay in each soil map unit; t is the thickness of each soil layer (Table 2); and depth to bedrock is the mean depth to bedrock for each map unit in centimetres.

The parent material class (Table 3) was derived from the STATSGO map unit component (comp) and taxonomic (tax) classification tables (USDA NRCS, 1995). The dominant mineralogy for each soil map unit was determined from the comp and tax tables. Each unit was classified into parent material class based on the mineralogical description (Table 3) (USDA NRCS, 2006).

Soil depth in meters was obtained from the CONUS-SOIL depth to bedrock layer. This layer identified map units with bedrock less than 1.52 m below the soil surface (i.e., map units coded with a depth of 1.52 m did not encounter bedrock) (Miller and White, 1998).

3.3. Forest area distribution

The USDA Forest Service, in collaboration with the US Geological Survey (USGS), developed a general forest cover

type data set (USDA Forest Service and US Geological Survey, 2000). Twenty-five forest type classes were interpreted from 1991 Advanced Very High Resolution Radiometer (AVHRR) and Landsat TM imagery. Twenty-one classes were used to divide forest cation uptake rates for the conterminous US. The four classes not used in the model represented lakes, non-forestland, non-US land, and ocean fill.

3.4. Wilderness areas

The National Atlas of the United States created a layer showing wilderness areas greater than 640 acres in the US. These wilderness areas are a part of the National Wilderness Preservation System of the US. Areas designated as wilderness are unmanaged and located on federal land (National Atlas of the United States, 2005).

3.5. Average annual runoff

A line coverage representing average annual runoff in inches per year in the US from 1951 to 1980 was produced by Gebert et al. (1987) data from over 5000 USGS streamflow gauging stations. The line coverage was converted into a 1-km² grid using the linegrid process in the GIS.

3.6. Base cation and nitrogen uptake

In the CAL SMBE, uptake refers to the N and base cations (i.e., Ca + Mg + K) lost through the removal of forest vegetation following periodic harvests. Uptake to support forest growth that remains on site is considered to be a form of internal

Table 3
Parent material class descriptions (modified from Gray and Murphy, 1999 to add parent material class column)

Parent material class	Parent material category	Silica content	Calcium–ferromagnesium content	Examples
Acidic	Extremely siliceous	>90%	Extremely low (generally <3%)	Quartz sands (beach, alluvial or Aeolian), chert, quartzite, quartz reefs and silicified rocks
Intermediate	Highly siliceous	72–90%	Low (generally 3–7%)	Granite, rhyolite, adamellite, dellenite, quartz sandstone and siliceous tuff
	Transitional siliceous/intermediate	62–72%	Moderately low (generally 7–14%)	Granodiorite, dacite, trachyte, syenite, most greywacke, most lithic sandstone, most argillaceous rocks and siliceous/intermediate tuff
	Intermediate	52–62%	Moderate (generally 14–20%)	Monzonite, trachy-andesite, diorite, andesite, intermediate tuff and some greywacke, lithic sandstone and argillaceous rock
Basic	Mafic	45–52%	High (generally 20–30%)	Gabbro, dolerite, basalt and mafic tuff (uncommon)
	Ultramafic	<45%	Very high (generally >30%)	Serpentinite, dunite, peridotite, amphibolite, and tremolite–chlorite–talcschists
	Calcareous	Low ^a	CaCO ₃ dominate other bases variable	Limestone, dolomite, calcareous shale and calcareous sands
Organic	Organic	Low ^a	Organic matter dominate bases variable	Peat, coal and humified vegetative matter
Other	Alluvial	Variable ^a	Variable	Variable
	Sesquioxide	Variable ^a	Variable, dominated by sesquioxides	Laterite, bauxite, ferruginous sandstone and ironstone

^a Category not defined by silica content.

cycling, rather than export. Nitrogen and base cation uptake were calculated using the following equation:

$$\text{Uptake}(\text{eq ha}^{-1} \text{ yr}^{-1}) = \text{AVI} \times \text{NC} \times \text{SG} \times \% \text{bark} \times 0.65 \quad (5)$$

where AVI is average forest volume increment ($\text{m}^3 \text{ ha}^{-1} \text{ yr}^{-1}$); NC is base cation or N nutrient concentration in bark and bole; SG is the specific gravity of bark and bole wood (g cm^{-3}); %bark is the percent of volume growth that is allotted to bark, and 65% of the tree volume is removed from the site (Birdsey, 1992; Martin et al., 1998; Hall et al., 1998).

Forest uptake of N, Ca, Mg, and K was calculated for bark and bole for all 21 classes in the forest type data set. Some classes were a mix of multiple species (i.e., red, white, and jack pine). The input parameters were split proportionally among the species to determine the input value for those classes. The uptake rates for both bark and bole were summed to estimate total annual base cation and nitrogen uptake.

Annual volume growth was calculated by region (North, South, Rocky Mountain, and Pacific Coast) and forest type using the FIA database. Approximately 15% of the deciduous volume growth and 11% of the coniferous volume growth can be allocated to aboveground bark (Wegner, 1984). The specific gravity for each species was obtained from a variety of references (Lamb and Marden, 1968; Erickson, 1972; Einsphar and Harder, 1976; Wegner, 1984; Hengst and Dawson, 1994). Nutrient concentrations for all base cations and N were obtained from the Tree Chemistry database, which reported nutrient concentration for foliage, bole bark, branches, bole wood, and twigs for tree species in the northeastern US (Pardo et al., 2004). When values for bark and bole were not available for species listed in the forest cover types, we used values of tree species with most comparable foliar nitrogen according to a previous foliar N meta-analysis (unpublished data). These data were used in Eq. (5) to estimate annual base cation and nitrogen uptake for all forest types (Table 4). These base cation and nitrogen uptake values are only relevant if wood is being removed from the forest. Areas designated as wilderness in the National Wilderness Preservation System of the US were given an uptake value of zero, because these areas are unmanaged and wood is not removed from the site.

3.7. Denitrification and nitrogen immobilization

Denitrification is the process by which nitrate is converted into gaseous N, most commonly in water saturated soil, and returned to the atmosphere. In the current SMBE, this parameter was set to zero to represent upland forests. Future applications of the SMBE will include the potential for delineating wetland forests, where significant denitrification rates have been recorded (Barton et al., 1999).

Nitrogen immobilization is the conversion of inorganic N to organic N. Gregor et al. (2004) reported values of nitrogen immobilization for forest soil plots ranging from 14.3 to 35.7 $\text{eq N ha}^{-1} \text{ yr}^{-1}$ in colder climates and up to 71.4 $\text{eq N ha}^{-1} \text{ yr}^{-1}$ in warmer climates. Because forest soil CAL was calculated for

Table 4
Base cation and nitrogen uptake values

Forest cover type	Nitrogen uptake ($\text{eq ha}^{-1} \text{ yr}^{-1}$)	Base cation uptake ($\text{eq ha}^{-1} \text{ yr}^{-1}$)
White-red-jack pine	59.07	77.14
Spruce fir	54.27	83.72
Longleaf slash pine	154.74	227.22
Loblolly shortleaf pine	140.41	208.58
Oak pine	129.71	213.75
Oak hickory	102.56	254.87
Oak-gum-cypress	124.18	235.68
Elm-ash-cottonwood	79.74	156.30
Maple-beech-birch	101.76	190.51
Aspen-birch	81.69	125.46
Douglas-fir	109.89	179.03
Hemlock-sitka-spruce	98.88	161.12
Ponderosa pine	75.29	174.39
Western white pine	40.69	37.11
Lodgepole pine	40.19	61.25
Larch	65.10	77.14
Fir-spruce	94.65	146.00
Redwood	100.92	156.62
Chaparral	106.60	201.61
Pinyon-juniper	40.87	58.21
Western Hardwoods	135.21	263.33

forests ranging from 45°N to 25°N, N immobilization was set to 42.86 $\text{eq N ha}^{-1} \text{ yr}^{-1}$ as an average of the colder and warmer climate N immobilization rates.

3.8. Acid neutralizing capacity

The critical leaching of forest soil acid neutralizing capacity (ANC) represents the buffering capacity of forest soils to acidic deposition and is therefore one of the most important determinants of a forest soil's CAL. Several factors can increase or decrease the critical ANC limit, as expressed in the following equation:

$$\text{ANC}_{(\text{le}, \text{crit})} = -Q^{2/3} \times \left(1.5 \times \frac{\text{BC}_{\text{dep}} + \text{BC}_{\text{w}} - \text{BC}_{\text{u}}}{K_{\text{gibb}} \times \left(\frac{\text{BC}}{\text{Al}} \right)_{\text{crit}}} \right)^{1/3} - 1.5 \times \frac{\text{BC}_{\text{dep}} + \text{BC}_{\text{w}} - \text{BC}_{\text{u}}}{\left(\frac{\text{BC}}{\text{Al}} \right)_{\text{crit}}} \quad (6)$$

where Q is annual runoff in $\text{m}^3 \text{ ha}^{-1} \text{ yr}^{-1}$; BC_{dep} is base cation (i.e., Ca + K + Mg) deposition; BC_{w} is forest soil base cation weathering; BC_{u} is base cation uptake by trees; K_{gibb} is the gibbsite equilibrium constant, a function of forest soil organic matter content (Table 5) that affects aluminum (Al) solubility (Gregor et al., 2004); and BC/Al is the assumed critical base cation to Al ratio, set at 1.0 for conifer forests (Gregor et al., 2004) and 10.0 for deciduous forests (Watmough et al., 2004). If the BC/Al ratio declines to values below the assumed critical limit, there is an increased likelihood of adverse impacts on trees.

Table 5
Gibbsite equilibrium constant as a function of forest soil organic matter content (modified from Gregor et al., 2004)

Soil type layer	Organic matter %	K_{gibb} (m^6/eq^2)
Mineral soils: C layer	<5	950
Soils with low organic matter: B/C layers	5–15	300
Soils with some organic material: A/E layers	15–30	100
Peaty and organic soils: organic layers	>70	9.5

4. Results

The spatial distributions of the forest soil CAL components as predicted by the SMBE were highly variable. The output of each SMBE component is discussed separately before CAL and exceedance estimates are presented.

4.1. Critical leaching of acid neutralizing capacity

Several factors influenced the forest soil critical leaching of ANC, including the K_{gibbsite} constant, which was partly a function of forest soil organic matter content. Soil organic matter content ranged from less than 5% over most of the US to over 30% in northern wetlands and eastern pocosin swamps (Fig. 1).

The critical ANC was also influenced by the base cation weathering rate (BC_w). A rough latitudinal gradient was found to be associated with base cation weathering (Fig. 2). Generally, northern and western forests had BC_w rates less than $2000 \text{ eq ha}^{-1} \text{ yr}^{-1}$, whereas the southeastern US generally

had BC_w rates between 500 and $3900 \text{ eq ha}^{-1} \text{ yr}^{-1}$. Florida and central California were notable exceptions with significant portions of those states having both lower and higher base cations weathering rates than the rest of their respective region.

The selection of the conifer BC/Al ratio of 1.0 and the deciduous BC/Al ratio of 10.0 also significantly influenced the calculated critical ANC limit of forest soils. The ANC limit of the forest soils across the US ranged from less than -500 to over $-4000 \text{ eq ha}^{-1} \text{ yr}^{-1}$, with the highest values generally found along the southeastern and northwestern US coasts (Fig. 3).

4.2. Deposition

Generally, S and N wet deposition rates decreased in a westerly direction, with rates in excess of $1300 \text{ eq ha}^{-1} \text{ yr}^{-1}$ in parts of New England (Fig. 4). Most western states had combined S and N deposition rates less than $200 \text{ eq ha}^{-1} \text{ yr}^{-1}$, with somewhat higher rates in southern California and western Washington. Florida, southern Carolina, southern Georgia, and Maine had combined S and N deposition rates below their regional averages.

Combined S and N dry deposition rates were largest in the Ohio, Pennsylvania, West Virginia, and eastern regions of the US, with rates exceeding $500 \text{ eq ha}^{-1} \text{ yr}^{-1}$. The southern US had S and N dry deposition rates ranging between 150 and $400 \text{ eq ha}^{-1} \text{ yr}^{-1}$. Within the southern region, dry deposition rates generally decreased in the western direction.

Only wet base cation (BC) deposition of Ca, Mg, Na, and K was included in the SMBE calculation of the forest soil CAL. The highest rates of BC deposition (not corrected for sea-salt)

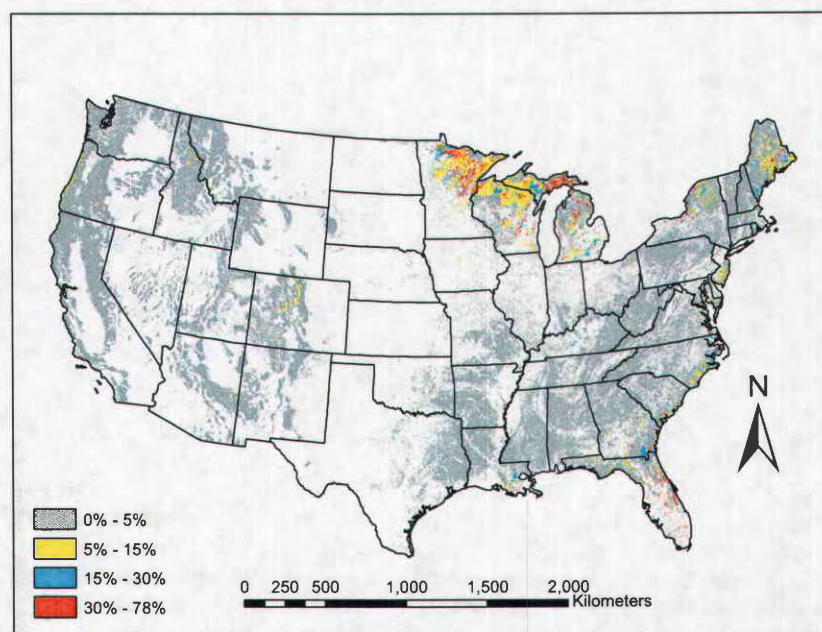


Fig. 1. Forest soil percent organic matter concentration averaged to a depth of 1.52 m for the conterminous US at a 1-km^2 spatial resolution.

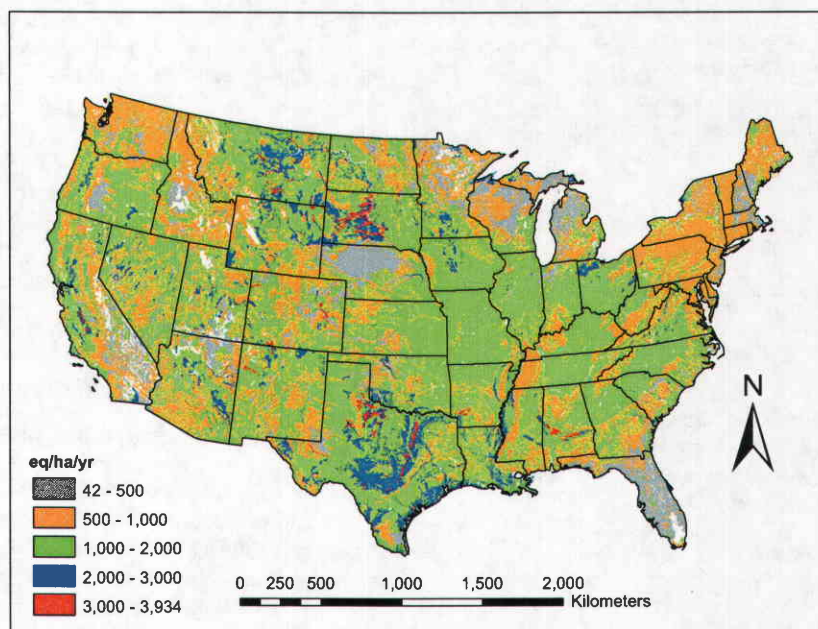


Fig. 2. Average annual forest soil base cation weathering expressed in $\text{eq ha}^{-1} \text{yr}^{-1}$ for the conterminous US for the years 1994–2000 at a 1-km^2 spatial resolution.

occurred along the northwestern and southeastern US coast and ranged from 200 to over $700 \text{ eq ha}^{-1} \text{yr}^{-1}$ (Fig. 5). The central Midwest had BC deposition rates ranging from 100 to $400 \text{ eq ha}^{-1} \text{yr}^{-1}$. The lowest BC deposition rates were found across much of the western US, where annual BC deposition was less than $100 \text{ eq ha}^{-1} \text{yr}^{-1}$.

Wet Cl deposition was included in the SMBE calculation of CAL. Given that Cl deposition is largely derived from ocean salts, Cl deposition was highest along the eastern and western US coasts. Annual Cl deposition rates ranged between 100 and $500 \text{ eq ha}^{-1} \text{yr}^{-1}$, and were lowest in the central US, with annual rates less than $100 \text{ eq ha}^{-1} \text{yr}^{-1}$ (Fig. 6).

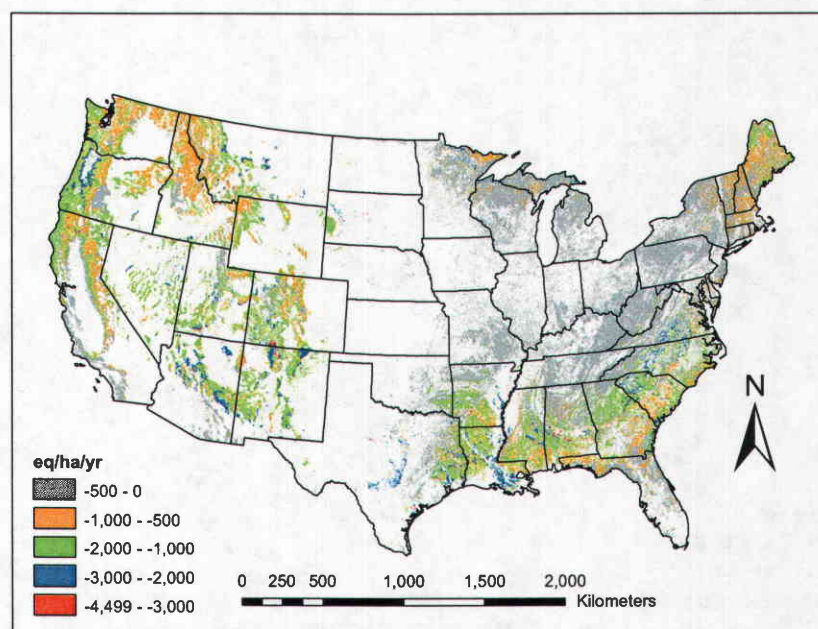


Fig. 3. Average annual critical leaching of forest soil acid neutralizing capacity expressed in $\text{eq ha}^{-1} \text{yr}^{-1}$ for the conterminous US for the years 1994–2000 at a 1-km^2 spatial resolution.

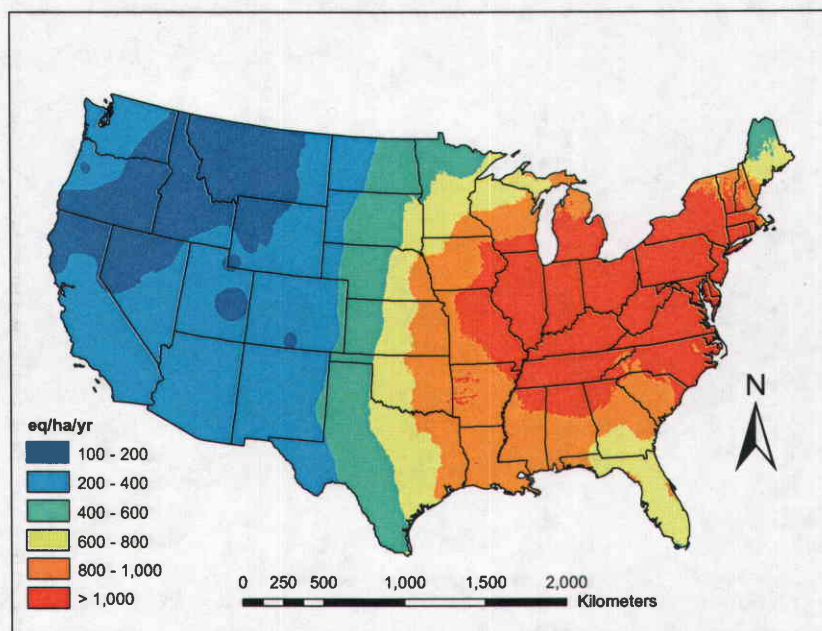


Fig. 4. Average annual combined wet and dry nitrogen and sulfur deposition expressed in $\text{eq ha}^{-1} \text{yr}^{-1}$ for the conterminous US for the years 1994–2000 at a 1-km^2 spatial resolution.

4.3. Critical acidic load

The predicted CAL in forest soils displayed a pattern similar to the critical ANC limit and BC_w . The highest forest soil CAL rates were found in the southeastern US, where CAL generally ranged from 1000 to over $8000 \text{ eq ha}^{-1} \text{yr}^{-1}$

(Fig. 7). However, much of the Appalachian Mountain range and Florida had CAL rates below $1000 \text{ eq ha}^{-1} \text{yr}^{-1}$.

Concern regarding the effects of acidic deposition on forest soil has focused largely on New England. However, the SMBE suggested that much of the Midwest and western regions of the US also have low forest soil CAL. In addition to low BC_w ,

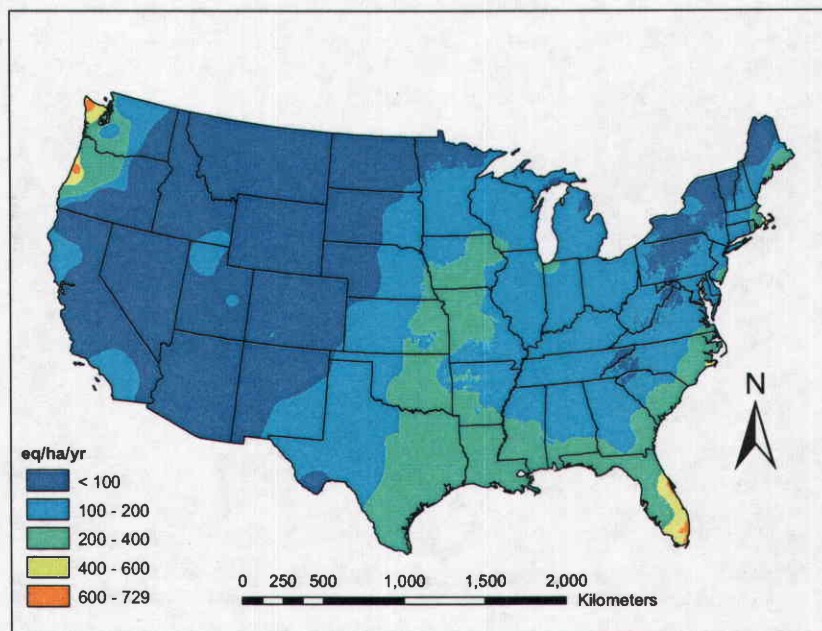


Fig. 5. Average annual base cation deposition expressed in $\text{eq ha}^{-1} \text{yr}^{-1}$ for the conterminous US for the years 1994–2000 at a 1-km^2 spatial resolution.

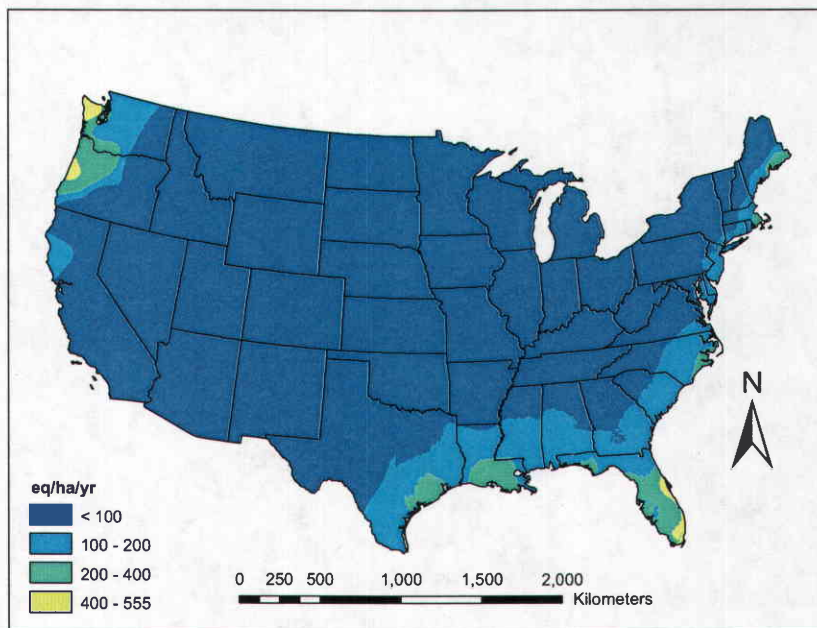


Fig. 6. Average annual wet chloride deposition expressed in $\text{eq ha}^{-1} \text{yr}^{-1}$ for the conterminous US for the years 1994–2000 at a 1-km^2 spatial resolution.

high rates of BC uptake by forests also contributed to a low CAL, particularly in the Midwest region.

4.4. Exceedance

Although the application of the SMBE did not have sufficient spatial resolution to differentiate exceedance within

a forest stand, regional patterns in forest soil CAL exceedance are evident. The SMBE suggested that much of the forest soil in New England and West Virginia is in exceedance of the CAL by over $500 \text{ eq ha}^{-1} \text{yr}^{-1}$ (Fig. 8). These are historic areas of concern for acid loading. The model also predicted that a small portion of southeastern North Carolina had forest soil CAL exceedances greater than $500 \text{ eq ha}^{-1} \text{yr}^{-1}$. No areas

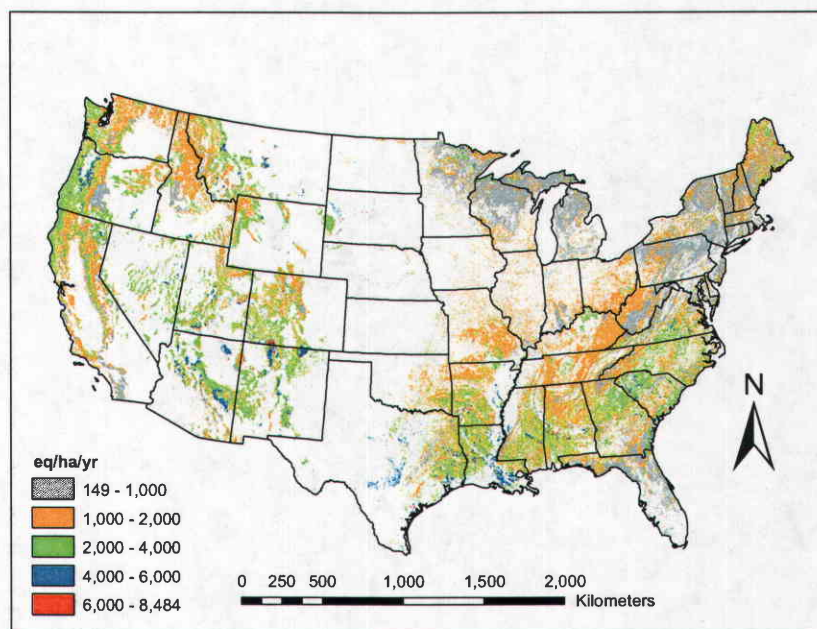


Fig. 7. Average annual critical acid load for forest soils expressed in $\text{eq ha}^{-1} \text{yr}^{-1}$ for the conterminous US for the years 1994–2000 at a 1-km^2 spatial resolution.

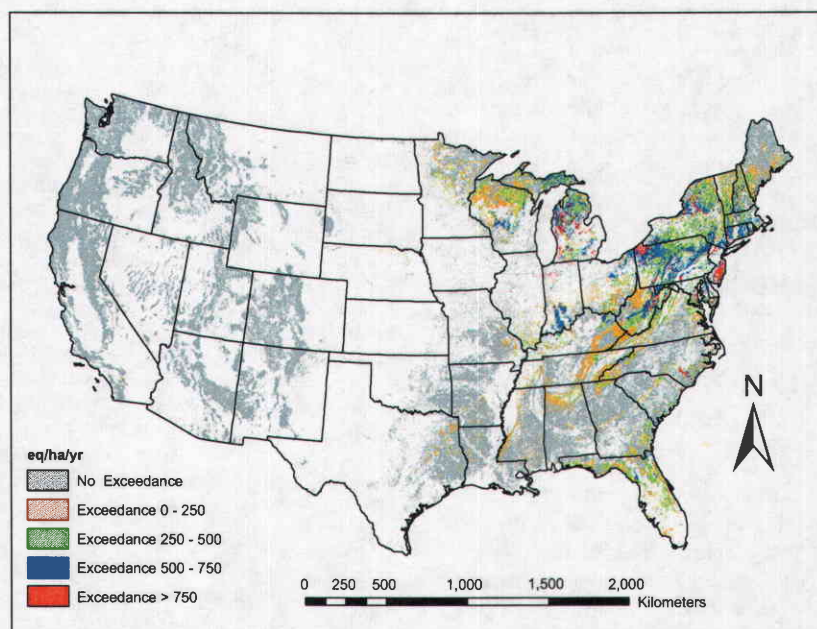


Fig. 8. Average annual exceedance of the critical acid load for forest soils expressed in $\text{eq ha}^{-1} \text{yr}^{-1}$ for the conterminous US for the years 1994–2000 at a 1-km^2 spatial resolution.

with forest soil CAL exceedance greater than $500 \text{ eq ha}^{-1} \text{yr}^{-1}$ were found in the western US, and only a few areas (e.g., southern California) showed any exceedance.

5. Discussion

5.1. Critical loads of acidity in forest soils

The SMBE predicted that 26% of US forest soils have CAL less than $1000 \text{ eq ha}^{-1} \text{yr}^{-1}$. Low CAL levels outside of New England, New York, and the Appalachian Mountain region are not necessarily problematic to forest health because acid deposition is much lower across most of the US compared to these areas. Unfortunately, mountainous terrain comprises much of the area with very low forest soil CAL. Mountain forests receive some of the highest local rates of acidic deposition. The combination of low forest soil CAL and high rates of acidic deposition is of special concern in some of these forests.

5.2. Annual exceedance of critical acid load

The SMBE predicted that average annual exceedance of forest soil CAL greater than $500 \text{ eq ha}^{-1} \text{yr}^{-1}$ represented approximately 7% of the total forest area across the conterminous US. In actuality, the amount of forest area in exceedance of the CAL by more than $500 \text{ eq ha}^{-1} \text{yr}^{-1}$ is probably higher, for several reasons. First, cloud deposition was not included in the estimates of total deposition. Second, base cation deposition to near-coastal areas was not corrected for marine aerosol contributions. Third, the 1-km^2 grid size averaged soils and deposition data, which removed extreme values from the estimation of forest floor CAL and exceedance. Locally (i.e., sub-km^2)

lower BC_w or higher acid deposition would be masked at a 1-km^2 resolution. Given these factors, the amount of forest soil with exceedance greater than $500 \text{ eq ha}^{-1} \text{yr}^{-1}$ may be significantly greater than that predicted by this SMBE.

The link between exceedance of the CAL and negative forest impacts has been observed in two eastern Canadian studies (Ouimet et al., 2001; Watmough et al., 2004). These studies did not find a causal relationship between exceedance of CAL and declining forest health, but the predicted distribution of forest soils with high CAL exceedance was consistent with research studies that cited negative forest impacts associated with high rates of N deposition across New England (Boggs et al., 2007; McNulty et al., 1991), and in West Virginia (Adams et al., 2006).

Outside the Northeast and West Virginia, forest soils in parts of the eastern North Carolina coastal plain were predicted to have a CAL exceedance greater than $500 \text{ eq ha}^{-1} \text{yr}^{-1}$. This part of North Carolina has very high rates of N deposition due to intensive hog farming (Aneja et al., 2001), and the predominantly sandy soils of the region have low base saturation (Fig. 3). Water quality monitoring by the USGS supports the SMBE predictions of forest soil CAL exceedance as stream nitrate concentrations in this area violate the national acceptable standard for drinking water (Spruill et al., 1998). The presence of high nitrate concentration in stream water is one of the impacts of CAL exceedance.

5.3. Limitations in using an SMBE to predict national-scale forest soil critical loads of acidity

These estimates of forest soil CAL did not account for acid deposition from clouds. Cloud water deposition can account for

35% (Miller et al., 1993) to 50% (Weathers et al., 2000) of total deposition at sites higher than 900 m in elevation. Although cloud deposition is clearly important in determining forest soils' CAL, current limitations associated with the projection of deposition across the landscape preclude their inclusion in this study. Also, SMBE can provide point-in-time estimates of forest soils' CAL and exceedance, but it cannot project how forest soil CAL may change over time. Forest tree composition, climate, soil acidity, and deposition patterns are not necessarily constant, and changes in CAL in response to these conditions cannot be calculated using an SMBE. Therefore, an SMBE cannot be used to estimate how long (if ever) it would take for an ecosystem to exceed its CAL.

6. Conclusions

An SMBE can provide a much-needed platform for comparing forest soil susceptibility to acidification across the conterminous US. Even though this study used a relatively coarse spatial scale (i.e., 1-km²) to aggregate data, the general patterns of forest soil acidity exceedance were still prominent. These initial findings should be considered preliminary. A more systematic analysis of model-predicted and measured forest soil CAL exceedance is needed before this approach can be used as a tool for identifying areas of potential forest health concern.

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References

- Adams, M.B., DeWalle, D.R., Hom, J.L. (Eds.), 2006. The Fernow Watershed Acidification Study. *Environmental Pollution Series*, vol. 11. Springer, New York, 279 pp.
- Aherne, J., Watmough, S., 2005. Critical loads of acidity for North America. In: *Acid Rain 2005, Seventh International Conference on Acid Deposition*, 12–17 June 2005, Prague, Czech Republic, 476 pp.
- Aneja, V.P., Roelle, P.A., Murray, G.C., Southerland, J., Erisman, J.W., Fowler, D., Asman, W.A.H., Patni, N., 2001. Atmospheric nitrogen compounds II: emissions, transport, transformation, deposition and assessment. *Atmospheric Environment* 35, 1903–1911.
- Barton, L., McLay, C.D.A., Schipper, L.A., Smith, C.T., 1999. Annual denitrification rates in agricultural and forest soils: a review. *Australian Journal of Soil Research* 37, 1073–1094.
- Baumgardner, Jr., R.E., Lavery, T.F., Rogers, C.M., Isil, S.S., 2002. Estimates of the atmospheric deposition of sulfur and nitrogen species: Clean Air Status and Trends Network 1990–2000. *Environmental Science & Technology* 36 (12), 2614–2629.
- Birdsey, R.A., 1992. Carbon Storage and Accumulation in United States Ecosystems. USDA Forest Service General Technical Report WO-59.
- Boggs, J.L., McNulty, S.G., Pardo, L.H., 2007. Changes in conifer and deciduous forest foliar and forest floor chemistry and basal area tree growth across a nitrogen (N) deposition gradient in the northeastern US. *Environmental Pollution* 149 (3), 304–314.
- Clean Air Act Amendments (CAAA) of 1990, 1990. Pub. L. No. 101-549, §7401-7671q, 104 Stat. 2399 (November 15, 1990).
- Cowling, E.B., 1981. An Historical Résumé of Progress in Scientific and Public Understanding of Acid Precipitation and Its Biological Consequences. SNSF Project Report Series, Oslo-Aas, Norway.
- Driscoll, C.T., Lawrence, G.B., Bulger, A.J., Butler, T.J., Cronan, C.S., Eagar, C., Lambert, K.F., Likens, G.E., Stoddard, J.L., Weathers, K.C., 2001. Acid rain revisited: advances in scientific understanding since the passage of the 1970 and 1990 Clean Air Act Amendments. In: Hubbard Brook Research Foundation, vol. 1. Science Links Publication. No. 1.
- Einsphar, D.W., Harder, M., 1976. Hardwood bark properties important to the manufacture of fiber products. *Forest Products Journal* 26 (6), 28–31.
- Erickson, J.R., 1972. The Moisture Content and Specific Gravity of the Bark and Wood of Northern Pulpwood Species. Research Note NC-141. USDA FS, North Central Forest Experiment Station.
- Gebert, W.A., Graczyk, D.J., Krug, W.R., 1987. Average Annual Runoff in the United States, 1951–80: U.S. Geological Survey Hydrologic Investigations Atlas HA-710, Scale 1:7,500,000.
- Gray, J.M., Murphy, B.W., 1999. Parent Material and Soils – A Guide to the Influence of Parent Material on Soil Distribution in Eastern Australia. Technical Report Number 45. NSW Department of Land and Water Conservation, Sydney, Australia.
- Grimm, J.W., Lynch, J.A., 2004. Enhanced wet deposition estimates using modeled precipitation inputs. *Environmental Monitoring and Assessment* 90, 243–268.
- Gregor, H.D., Werner, B., Spranger, T. (Eds.), 2004. Manual on Methodologies and Criteria for Mapping Critical Levels/Loads and Geographical Areas Where They are Exceeded. ICP Modeling and Mapping. Umweltbundesamt, Berlin, Germany, 212 pp.
- Hall, J., Bull, K., Bradley, I., Curtis, C., Freer-Smith, P., Hornung, M., Howard, D., Langan, S., Loveland, P., Reynolds, B., Ullyett, J., Warr, T., 1998. Status of UK Critical Loads and Exceedances. Part 1—Critical Loads and Critical Loads Maps. Report to the Department of Environment under DETR/NERC Contract EPG1/3/116. Available from: <<http://critloads.ceh.ac.uk>>.
- Hall, J., Hornung, M., Kennedy, F., Langan, S., Reynolds, B., Aherne, J., 2001. Investigating the uncertainties in the simple mass balance equation for acidity critical loads for terrestrial ecosystems. *Water, Air, and Soil Pollution: Focus* 1, 43–56.
- Hengst, G., Dawson, J.O., 1994. Bark properties and fire resistance of selected tree species from the central hardwood region of North America. *Canadian Journal of Forest Research* 24, 688–696.
- Lamb, F.M., Marden, R.M., 1968. Bark specific gravities of selected Minnesota tree species. *Forest Products Journal* 18 (9), 76–82.
- Martin, J.G., Kloepfel, B.D., Schaefer, T.L., Kimbler, D.L., McNulty, S.G., 1998. Aboveground biomass and nitrogen allocation of ten deciduous southern Appalachian tree species. *Canadian Journal of Forest Research* 28, 1648–1659.
- McNulty, S.G., Aber, J.D., Boone, R.D., 1991. Spatial changes in forest floor and foliar chemistry in spruce–fir forests across New England. *Biogeochemistry* 14, 13–26.
- Miller, D.A., White, R.A., 1998. A conterminous United States multi-layer soil characteristics data set for regional climate and hydrology modeling. *Earth Interactions* 2. Available from: <<http://earthinteractions.org>>.
- Miller, E.K., Panek, J.A., Friedland, A.J., Kadlec, J., Mohnen, V.A., 1993. Atmospheric deposition to a high-elevation forest at Whiteface Mountain, New York, USA. *Tellus* 45B, 209–227.
- Minami, M., 2000. Using ArcMap. Environmental Systems Research Institute, Inc., Redlands, CA, 528 pp.
- National Atlas of the United States, 2005. December 2005. National Wilderness Preservation System of the United States. National Atlas of the United States, Reston, Va. Available from: <<http://nationalatlas.gov>>.
- National Atmospheric Deposition Program (NADP) (NRSP-3), 2005. Illinois State Water Survey. NADP Program Office, Champaign, IL. Available from: <<http://nadp.sws.uiuc.edu/isopleths/>>.

- Nilsson, J., Grennfelt, P.-I. (Eds.), 1988. Critical Loads for Sulphur and Nitrogen, vol. 15. Nordic Council of Ministers, Miljørapport. 418 pp.
- Ouimet, R., Duchesne, L., Houle, D., Arp, P.A., 2001. Critical loads of atmospheric S and N deposition and current exceedances for northern temperate and boreal forests in Quebec. *Water, Air, and Soil Pollution: Focus* 1 (1–2), 119–134.
- Pardo, L.H., Robin-Abbott, M., Duarte, N., Miller, E.K., 2004. Tree Chemistry Database (Version 1.0). General Technical Report NE-324. US Department of Agriculture, Forest Service, Northeastern Research Station, Newton Square, PA, 45 pp.
- Posch, M., Hettelingh, J.-P., Mayerhofer, P., 2001. Past and future exceedances of nitrogen critical loads in Europe. *The Scientific World* 1 (s2), 945–952.
- Spruill, T.B., Harned, D.A., Ruhl, P.M., Eimers, J.L., McMahon, G., Smith, K.E., Galeone, D.R., Woodside, M.D., 1998. Water Quality in the Albemarle-Pamlico Drainage Basin, North Carolina and Virginia, 1992–95. US Geological Survey Circular 1157, 36 pp.
- Sverdrup, H., de Vries, W., Henriksen, A., 1990. Mapping Critical Loads: A Guidance to the Criteria, Calculations, Data Collection and Mapping of the Critical Loads. Miljørapport (Environmental Report) 1990: 14. Nordic Council of Ministers, Copenhagen, NORD: 1990: 98, 124 pp.
- Sverdrup, H., de Vries, W., 1994. Calculating critical loads for acidity with the simple mass balance equation. *Water, Air, and Soil Pollution* 72, 143–162.
- US Environmental Protection Agency (EPA), 2006. Clean Air Status and Trends Network (CASTNET). Available from: <<http://www.epa.gov/castnet/>>.
- US Environmental Protection Agency (EPA), 2007. Clean Air Markets: Data and Maps. Available from: <<http://cfpub.epa.gov/gdm/index.cfm?fuseaction=aciddeposition.wizard>>.
- USDA Forest Service (USFS) and US Geological Survey (USGS), 2000. Forest Cover Types. US Geological Survey, Reston, VA. Available from: <<http://nationalatlas.gov/metadata/foresti0201.xml>>.
- USDA Natural Resources Conservation Service (NRCS), 1995. State Soil Geographic (STATSGO) Data Base: Data Use Information. Miscellaneous Publication Number 1492. National Soil Survey Center, 113 pp. Available from: <<http://www.aces.edu/waterquality/data/soils/statsgo/statsgo-user-guide.pdf>>.
- USDA Natural Resources Conservation Service (NRCS), 2006. Keys to Soil Taxonomy, 10th ed. Soil Survey Staff, 332 pp.
- Watmough, S.A., Aherne, J., Dillion, P.J., 2004. Critical Loads Ontario: Relating Exceedance of the Critical Loads with Biological Effects at Ontario Forests. Report 2, Environmental and Resource Studies. Trent University, Ontario, 22 pp.
- Weathers, K.C., Lovett, G.M., Likens, G.E., Lathrop, R., 2000. The effect of landscape features on deposition to Hunter Mountain. Catskill Mountains, New York. *Ecological Applications* 10 (2), 528–540.
- Wegner, K.F., 1984. *Forestry Handbook*, second ed. John Wiley & Sons Inc., New York, 1335 pp.
- Wright, R.F., Schindler, D.W., 1995. Interactions of acid rain and global change effects on terrestrial and aquatic ecosystems. *Water, Air, and Soil Pollution* 85, 89–99.

