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Susceptibility of Forests in the Northeastern USA to Nitrogen and Sulfur Deposition: Critical Load Exceedance and Forest Health

N. Duarte · L. H. Pardo · M. J. Robin-Abbott

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Abstract The objectives of this study were to assess susceptibility to acidification and nitrogen (N) saturation caused by atmospheric deposition to northeastern US forests, evaluate the benefits and shortcomings of making critical load assessments using regional data, and assess the relationship between expected risk (exceedance) and forest health. We calculated the critical loads of nutrient N and of sulfur (S)+N using the steady-state mass balance method at >4,000 regional and national vegetation and soil monitoring network plots in the northeastern USA. Regional calculations of critical loads necessitate use of soil maps which provide a range for each soil characteristic resulting in a broad range of critical load of S+N and exceedance values. For the scenario most representative of regional conditions, over 80 % of the critical loads fell into the range of 850–2050 eq ha^{-1}yr^{-1}; at 45 % of the plots, deposition exceeded the critical load. In contrast, the critical load for nutrient N, 200–300 eq ha^{-1}yr^{-1}, was lower. Site measurements, especially to estimate soil weathering, would increase the certainty of the critical load. We observed significant negative correlations between critical load exceedance and growth (17 species) and crown density (4 species); we observed significant positive correlations of exceedance with declining vigor (four species), with crown dieback (six species) and crown transparency (seven species). Among the species which demonstrate the most significant detrimental responses to atmospheric deposition are balsam fir, red spruce, quaking aspen, and paper birch. These results indicate that significant detrimental responses to atmospheric deposition are being observed across the northeastern USA.

Keywords Acidification · Base cation depletion · Forest health · N saturation · Simple mass balance model

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1 Introduction

Anthropogenic activities caused emissions of sulfur (S) and nitrogen (N) to increase dramatically in the middle of the twentieth century (Driscoll et al. 2001). Although S emissions have since decreased significantly as a result of SO_2 control programs (Driscoll et al. 2001), projected emissions of acidifying S and N compounds are expected to have continuing negative impacts on forests. Atmospheric S and N deposition have contributed to acidification of soils and surface waters, export of nutrient cations, and mobilization of aluminum (Al) in soils (Reuss 1983; Reuss and Johnson 1985), which can be toxic to plants and other biota. When exports of nutrient cations are greater than inputs to an ecosystem, soil nutrients may decrease to inadequate levels, a condition known as cation depletion. Cation depletion may result in a wide range of forest health problems: reduced growth rates and increased susceptibility of forests to climate change; pest and pathogen stress, which results in reduced forest health, reduced timber yield, increased mortality; and eventual changes in forest species composition (Schaberg et al. 2001; Bailey et al. 2005). In addition to these acidification impacts, excess N deposition can lead to N saturation, the condition when N inputs exceed biotic demand (Aber et al. 1989, 1998). Excess N may result not only in elevated nitrate leaching and further stream and soil acidification but may also lead to plant nutrient imbalances, which ultimately lead to similar forest health problems as cation depletion (Pardo et al. 2011c). In northeastern North America, where N and S deposition are relatively high (NADP 2009), these N and S emissions, therefore, present long-term threats to forest health and productivity.

Critical loads have been used as a tool in the process of negotiating decreases in air pollution in Europe (Posch et al. 1995, 2001). The critical load is the level of deposition below which no harmful ecological effects occur for a forest ecosystem over the long term (Nilsson and Grennfelt 1988). Exceedance is the difference between the current deposition and the critical load (UBA 2004). In the USA, critical loads are beginning to be widely used within federal agencies (Fenn et al. 2011). Because of the intensive data demands, the scope of critical loads estimates has often been limited. Using simple models to extrapolate data allows estimation of critical loads for many locations in the northeastern USA; however, using regional-scale data lowers the certainty of critical load estimates. This assessment provides resource managers with information to enable them to evaluate the quality of regional critical load estimates and to identify species at risk from atmospheric deposition.

The objectives of this study were to: (1) assess forest susceptibility to atmospheric deposition in the northeastern USA by calculating the critical load of S+N and of nutrient N and exceedance, (2) evaluate the benefits and shortcomings of making CL assessments using regional data, and (3) relate measures of forest susceptibility to atmospheric deposition with indicators of forest health by comparing ecological indicators (crown health and growth) at the plot level to exceedance.

2 Materials and Methods

This study, which focused on using datasets available on the regional scale that could be adapted for use in critical loads calculations, contributed to a larger forest sensitivity mapping project for New England and Eastern Canada. The larger project was an initiative of the New England Governors/Eastern Canadian Premiers (NEG/ECP) to map forests sensitive to atmospheric deposition in New England and Eastern Canadian (NEG/ECP Forest Mapping Group 2001, 2003).

2.1 Site Description

We included 4,057 plots from national and regional forest health surveys in this analysis (Fig. 1 and Table 1). These included the plots from the national USDA Forest Service Forest Inventory and Analysis (FIA) program, including the Forest Health Monitoring (FHM) program (Coulston et al. 2005; USDA-FS 2006). The FIA program's Forest Monitoring Survey is described in detail at: http://www.fia.fs.fed.us; further details on sites can be found in Duarte et al. (2011a) and the Electronic Supplementary Materials (ESM). Other forest health surveys in New England states, including the North American Maple Program (NAMP), the Vermont Hardwood Health Survey (HHS), and the Vermont Monitoring Cooperative Forest Health Plots (VMC-FH), were included in this assessment. Data from the National Resource Inventory soil pits and from county soil surveys (Natural Resource Conservation Service (NRCS) Soil Survey Staff 2003) and additional research sites in Vermont were also included (Table 2).

The sites based on county soil surveys in the NRCS National Soil Survey Center (NSSC) database (Soil Survey Staff 2003) were defined as forested by the presence of an organic layer or, in some cases, the

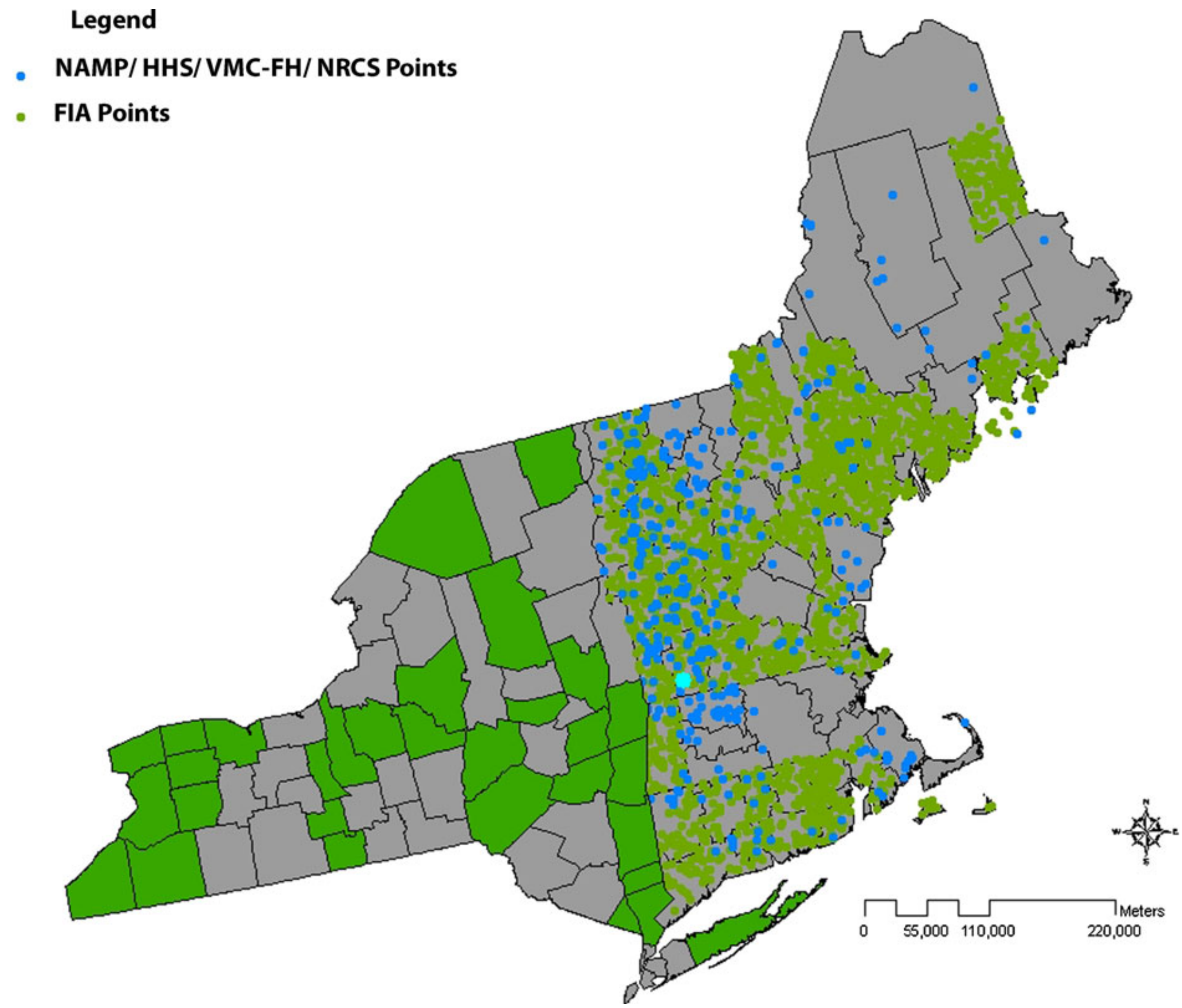

Fig. 1 Site location New England and New York. Only sites for which digitized soil maps were available are included in this analysis. These include plots from the North American Maple Program (NAMP), the Vermont Hardwood Health Survey (HHS), and the Vermont Monitoring Cooperative Forest Health Plots (VMC-FH) and Natural Resources Conservation Service soil surveys (NRCS). US Forest Service Forest Inventory and Analysis (FIA) P2 plots are indicated using the publicly available (not true) coordinates (see Section 2.3). Because only county center coordinates are publicly available for New York, counties where digitized soil maps are available are indicated in *blue*. FHM (P3) plots do not have publicly available coordinates and are not shown on this map

absence of an Ap horizon combined with a record of forest vegetation. Forested pedons with evidence of gleying (indicating wet soil) were excluded from our analysis. The additional research sites in Vermont include plots for which sufficient data were available to estimate critical loads. These include plots on Camel's Hump, Mount Ascutney, and 75 other plots throughout the state (C. Cogbill, unpublished data). Species composition and some limited soil data (parent material; soil series; in some cases, soil depth) were available for the additional research sites. Only sites for which digitized soil maps were available are included in this analysis (Tables 1 and 2; Fig 1), which limited the extent of coverage.

2.2 Critical Load Calculations

2.2.1 Critical Load for Acidity (S+ N)

Calculations of critical loads of S+N are based on a simple mass balance model described in detail elsewhere (UBA 2004; Pardo 2010:

Table 1 Distribution of sites by state

Site group	CT	MA	ME	NH	NY	RI	VT	Total
FIA (P2 plots)	231	116	631	552	1,012	99	509	3,150
FHM (P3 plots)	12	3	31	22	402	4	17	491
VMC-FH							19	19
NAMP	10	10	18				35	73
VT HHS							62	62
NRCS	27	56	33	37		2	61	216
VT additional research sites							75	75
Total	280	185	713	591	1,414	103	778	4,064

$$\text{Critical load}\,(S+N) = BC_{dep} + BC_w - Bc_u + N_i + N_u + N_{de} - ANCle_{crit} \qquad (1)$$

Where:

BC_{dep} sum of Ca+Mg+Na+K deposition rate (eq $ha^{-1}yr^{-1}$)
BC_w soil weathering rate of Ca+Mg+K+Na (eq $ha^{-1}yr^{-1}$)
Bc_u net Ca+Mg+K uptake rate (eq $ha^{-1}yr^{-1}$) removed by harvest or disturbance
N_i acceptable immobilization (accumulation) of N in soil
N_u N uptake (removal of N by harvest or fire)
N_{de} export of N via denitrification
$ANC_{le(crit)}$ acceptable acid neutralizing capacity (ANC) leaching rate (eq $ha^{-1}yr^{-1}$).

The acceptable ANC leaching rate, $ANC_{le(crit)}$, is calculated based on the critical chemical criteria of no change in base saturation according the NEG/ECP Forest Mapping Group Protocol (NEG/ECP 2001). In order to achieve the condition of no change in base saturation, a BC/Al_{crit} ratio of 10 (moles per mole) was used (NEG/ECP 2001; Ouimet et al. 2006).

$$ANC_{le\,(crit)} = -Q^{\frac{2}{3}} \times \left(1.5 \times \frac{BC_{dep} + BC_W - BC_u}{K_{Gibb} \times \left(\frac{BC}{Al_{crit}}\right)}\right)^{\frac{1}{3}} - 1.5 \times \frac{BC_{dep} + BC_W - BC_u}{\frac{BC}{Al_{crit}}} \qquad (2)$$

Where:

Q precipitation surplus or streamflow (cubic meters per hectare per year)
K_{Gibb} 10^9 (moles per liter)^2

Table 2 Site parameters

Study	Location	Plot size (ha)	Soil data	Vegetation data	Forest health data
FIA (P2 plots)	CT, MA, ME, NH, NY, RI, VT	0.067		Species level	Growth
FHM (P3 plots)	CT, MA, ME, NH, NY, RI, VT	0.067		Species level	Crown density and dieback; canopy transparency
VMC-FH	VT	0.067		Species level	Crown density and dieback; canopy transparency
NAMP	CT, MA, ME, VT	0.01		Species level	Vigor, canopy transparency, crown dieback
VT HHS	VT	1		Species level	Vigor, canopy transparency, crown dieback
NRCS	CT, MA, ME, NH, RI, VT		Soil pedon data	No data	No data
VT additional research	VT		Limited site data	Species level	No data

2.2.2 Critical Load for Nutrient N

We also calculated the critical load with respect to nutrient N, which is the rate of N deposition below which nutrient imbalances or other detrimental consequences of N deposition do not occur (NEG/ECP 2001; UBA 2004). The critical load for nutrient N is the level of deposition that would balance the acceptable accumulation and export of N in the forest ecosystem.

$CL_{nut}(N)$ can then be expressed as:

$$CL_{nut}(N) = N_i + N_u + N_{de} + N_{le(acc)} \quad (3)$$

where:

$N_{le(acc)}$ acceptable nitrate leaching

The acceptable net N accumulation (N_i) is not a measured value, but is set based on the accumulation of N in soil that would be considered unlikely to lead to or signal disruptions in the N cycle (UBA 2004; Pardo 2010). In these calculations, we used an acceptable soil N accumulation rate of 2 kgha^{-1}year^{-1}. There is little research in the USA upon which to base the values for acceptable soil N accumulation and acceptable nitrate leaching; even the established CL approach in Europe notes that there is no consensus on sustainable long-term rates of soil N accumulation (UBA 2004) The calculation of net N uptake is described in Section 2.3.2, the ESM, and Duarte et al. (2011a). For upland forest soils, denitrification rates are small to negligible (Seitzinger et al. 2006); hence, denitrification is set to 0. The acceptable nitrate leaching rate, $N_{le(acc)}$, is the maximum acceptable leaching rate for an ecosystem that is not at N saturation. This leaching rate is given by:

$$N_{le(acc)} = Q \times [N]_{crit} \quad (4)$$

where:

$[N]_{crit}$ the nitrate concentration in the soil solution above which it would be considered detrimental to ecosystem or soil

$[N]_{crit}$ was set at 0.2 g Nm^{-3} (de Vries et al. 2007; UBA 2004).

2.3 Input Data

Both measured and modeled data were used as inputs for deposition, climate, vegetation, and soil parameters (Duarte et al. 2011a, b; NEG/ECP 2001). In order to display the results spatially for FIA plots while protecting landowner privacy, we used publically available "fuzzed and swapped" co-ordinates that were switched with those for a similar plot within the county (swapped) and altered by approximately 0.5 miles (fuzzed). These changes should not alter general patterns at the regional scale.

2.3.1 Climate and Deposition

We used the ClimCalc model to calculate the required climate and deposition parameters (Ollinger et al. 1993; http://www.pnet.sr.unh.edu/climcalc). Climate values are used to calculate the soil mineral weathering rate. Base cation deposition data are used to calculate the critical load; N and S deposition data are used to calculate the exceedance. Monthly precipitation volume and air temperature were combined with soil attributes in a simple model that estimates annual evapotranspiration (Miller personal communication; see Miller 2011 for details).

Precipitation volume across New England and New York ranges from 64 to 209 cm (mean, 112 cm). Annual evapotranspiration ranges from 3 to 74 cm (mean, 33 cm). Sulfur deposition ranges from 242–1,154 eq ha^{-1}yr^{-1} (~4–18 kg S ha^{-1}year^{-1}); N deposition ranges from 256–920 eq ha^{-1}yr^{-1} (~4–13 kgN ha^{-1}year^{-1}), with higher deposition rates in New York and southwestern New England (Fig. 2). Base cation deposition rates range from 62 to 286 eq ha^{-1}yr^{-1}.

2.3.2 Nutrient Removal

Removal of biomass from forest ecosystems by harvesting or fire results in the removal of nutrient base cations and N. As fire is not significant in this region (Richburg and Patterson 2000), we calculated only nutrient removal via harvesting. All of the required data for calculating annual merchantable removals of growing stock trees on timberland are available in the publicly accessible FIA database (http://www.fia.fs.fed.us/tools-data). Current removal rates at the county level (FIA Database Documentation) were combined with chemistry data for species found in the northeastern USA from the Tree Chemistry Database (Pardo et al. 2005) in order to calculate the annual nutrient removal rates. FIA plot data were collected between 1993 and 2004 (see Duarte et al. 2011a). For non-FIA

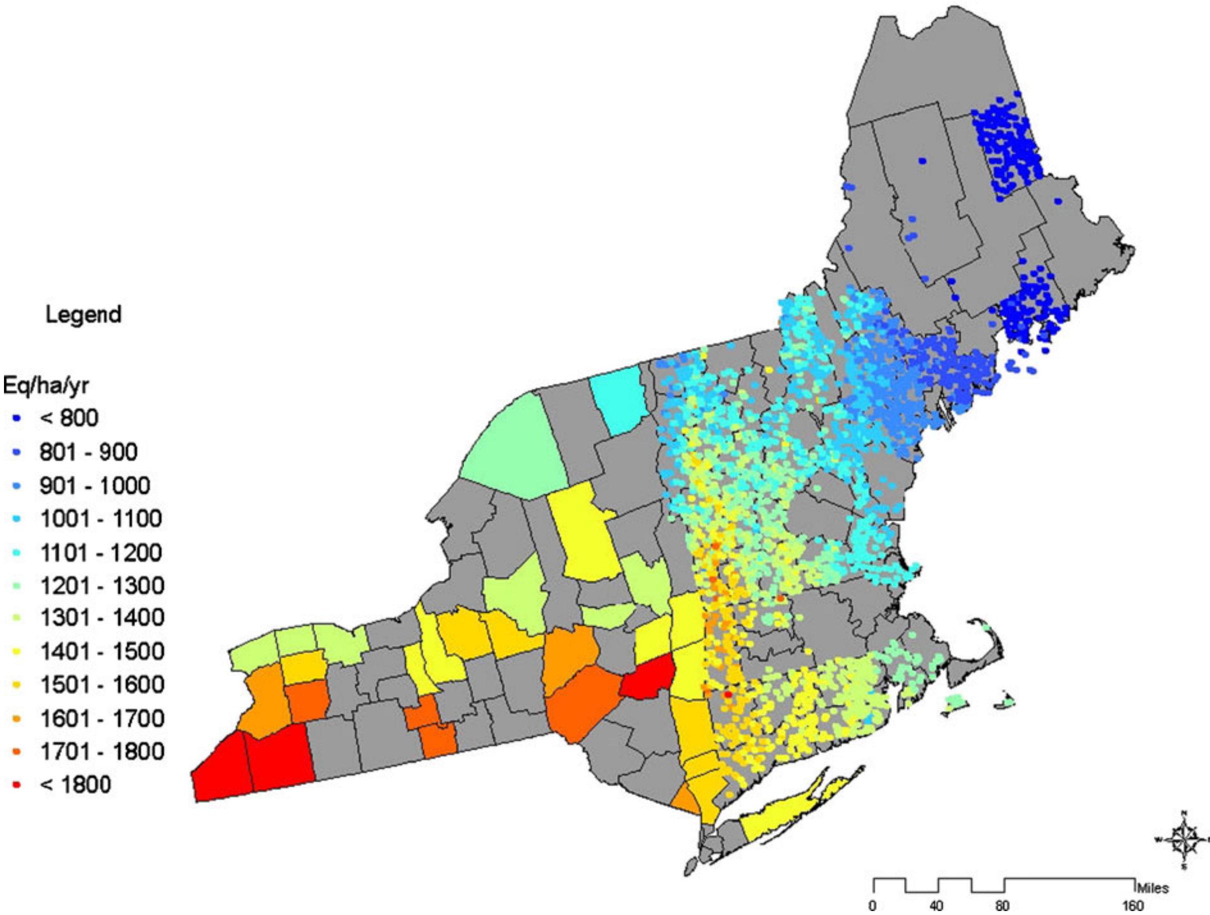

Fig. 2 S+N deposition rates (wet+dry) modeled by ClimCalc for sites in New England and New York (eq ha^{-1}yr^{-1})

sites, we assumed saw timber harvest to be the dominant harvest type and used species composition and DBH data to calculate tree wood and bark biomass removed using allometric equations (Jenkins et al. 2003; see Duarte et al. 2011a). Annual biomass extraction rates for saw timber compiled from FIA data and tabulated by county, land-ownership category (public and private), and gross forest type (softwood, hardwood, mixed) were used to estimate the nutrient removal from these non-FIA sites, except the NRCS sites. For the NRCS sites, which have no vegetation data, we assumed no biomass removal.

2.3.3 Soil Mineral Weathering Rates

Mineral weathering represents the most significant term in the critical loads calculations. Soil depth, texture, and moisture data were used with monthly precipitation volume and air temperature to model annual evapotranspiration. Soil depth, clay percent, substrate type (see Duarte et al. 2011b for substrate classification of soil series), and mean annual air temperature were used to estimate mineral weathering rates using the clay percent/substrate type method (Sverdrup et al. 1990; Ouimet et al. 2006; McNulty et al. 2007; see ESM for calculation method). The clay percent/substrate type method uses three categories of soil substrate: acidic, intermediate, and basic. Acidic soil substrates include granites, gneiss, sandstones, and felsic rocks; intermediate soil substrates include diorite, granodiorite, conglomerates, and most sedimentary rocks other than sand stone; basic soil substrates include mafic rocks, sedimentary rocks with low carbonate content, and carbonate rocks. Estimates for weathering rates were made for the rooting zone; when we had information that indicated a root limiting layer, we included only horizons above that layer.

In order to identify the primary soil series for each plot, the FIA GIS Specialist overlaid the true geographic coordinates of the FIA and FHM plots on the digitized soil county survey maps (Soil Survey Geographic (SSURGO) Database; http://soils.usda.gov/survey/geography/ssurgo). Regions for which digitized soil maps were not available were excluded from this analysis (Fig. 1). We created a database of parameters associated with each soil series, based on NSSC soil pit data (Soil Survey Staff 2003) and Official Soil Series Description (OSSD). The OSSD were used to determine minimum, maximum, and midpoint values for the required soil parameters (depth, clay percent, texture, moisture, and substrate type). In order to reduce an unrealistic skewing of the minimum weathering rate towards zero, we eliminated the lowest 20 % of the range of values reported in the OSSD and used this adjusted value as the minimum weathering rate. The "mid" value was calculated by taking the midpoint of the range ($\text{maximum}_{\text{reported}} - \text{minimum}_{\text{adjusted}}$) of OSSD parameters for each soil type. The NSSC soil pit data (Soil Survey Staff 2003) were averaged across New England to generate mean values for the soil input parameters by soil series. Our objective in taking the mean of the NCCS soil pit data was to constrain the range of possible values (from the OSSD) to better reflect the range of what is actually observed in the region. However, the number of pits per soil series and state was so limited that it was difficult to assess how representative these plots were. Nonetheless, we report NSSC mean weathering rates for most sites (see ESM Fig. S1). For some soil series, soil data were not available from the NSSC soil database. For plots with these soil series, only minimum, maximum, and midpoint mineral weathering rates were calculated (based on the OSSD). Mineral weathering rates are shown by state for all sites (Fig. 3).

2.4 Ecological Indicators of Forest Health

Ecosystem health and vitality were measured using tree health indicators: canopy transparency, crown density, vigor, dieback, and growth. These ecological indicators were employed to assess the current health status at each site and were then compared with the exceedance by species. We excluded trees with damage caused by logging. We analyzed FIA P3, HHS, NAMP, and VMC-FH data from 1995. We analyzed FIA P2 data from inventory years 1994 to 1998 (Duarte et al. 2011a).

The ecological indicator data included in our analysis are from tree measurements at individual ground plots. We included only measurements made for dominant, co-dominant, and open grown trees in our analysis because these trees are less likely to decline due to confounding stand dynamics and therefore more easily interpreted for forest health decline related to atmospheric deposition. We compared critical load exceedance with forest health indicators using the Spearman's rank correlation analysis (α=0.05) using SAS software.

3 Results

The susceptibility of forest ecosystems to negative impacts from atmospheric deposition was assessed by making calculations of critical load for acidity (S+N) and for nutrient N (N_{nut}). Sites where the deposition rate exceeds the critical load are considered susceptible to negative impacts from N and S deposition. The critical load, and therefore the exceedance, varied depending on the weathering rate scenario used, resulting in worst case (using minimum weathering rates), mid, best case (using maximum weathering rates), and NSSC mean scenarios for each site.

3.1 CL(S+N)

The critical load tended to be lowest in northern New England, with higher values reported in southern New England, coastal areas, and parts of New York. For example, in Connecticut and Rhode Island, most critical loads using the mid-point weathering rates (~80 %) fell in the range of 1,500–2,000 eq ha^{-1} yr^{-1}; while in Maine, New Hampshire, and Vermont, ~65 % of critical loads were less than 1,500 eq ha^{-1} yr^{-1} (Fig. 4a).

In order to evaluate patterns in the critical loads, we determined the percentage of plots that fell between the lower and upper inflection points on the cumulative frequency plots (Fig. 6a). This represents the majority of the sites in each state and eliminates the tails of the distribution, which can be quite broad.

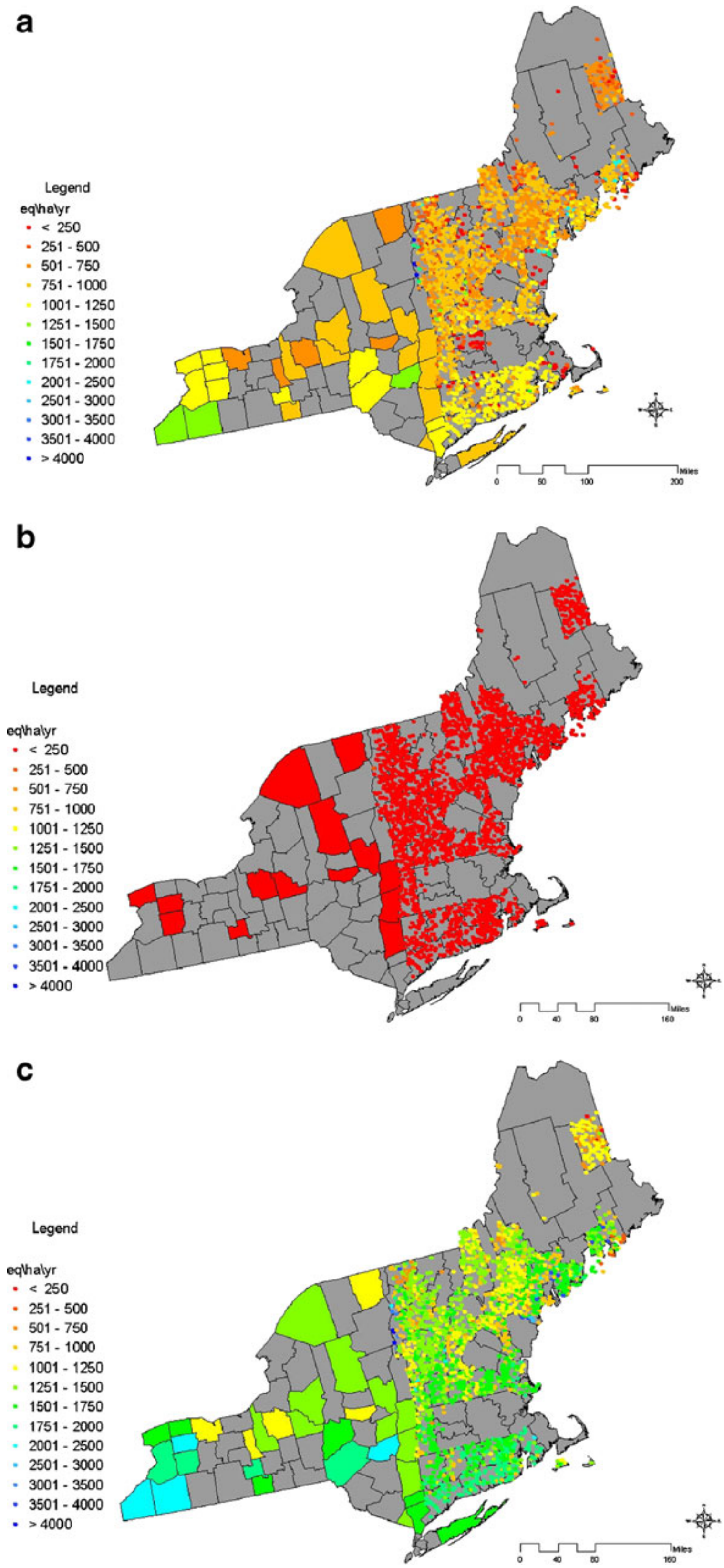

Fig. 3 Weathering rates for sites in New England and New York. **a** Midpoint scenario. Midpoint of the range of minimum and maximum weathering rates calculated based on clay percent and substrate type using Official Soils Series descriptions and county soil surveys (described in Section 2.3.3). **b** Worst case scenario. Minimum weathering rate calculated based on clay percent and substrate type using Official Soils Series descriptions and county soil surveys (described in Section 2.3.3). **c** Best case scenario. Maximum mineral weathering rates calculated based on clay percent and substrate type using Official Soils Series descriptions and county soil surveys (described in Section 2.3.3)

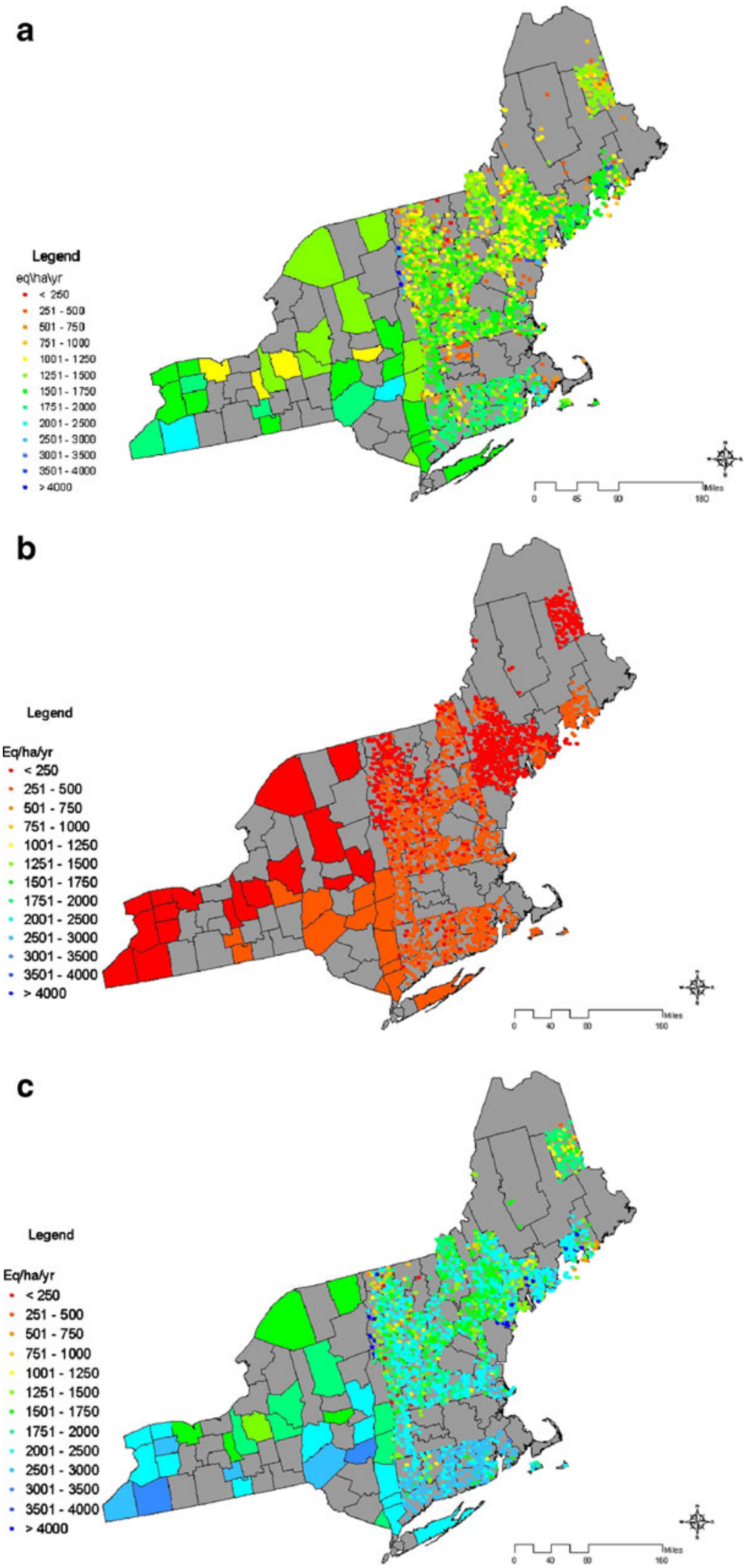

Fig. 4 Critical loads for acidity (S+N) for sites in New England and New York. **a** Midpoint scenario. **b** Worst case scenario. **c** Best case scenario

Critical loads calculated using the mid-point weathering rates mostly (>80 %) fell between 850 and 2,050 eq ha^{-1}yr^{-1}, the total range was 11 to 6,540 eq ha^{-1}yr^{-1} (Figs. 4a and 6a). Critical loads for the worst case scenario, calculated using the minimum weathering rates, were considerably lower, with 90 % falling below 1,085 eq ha^{-1}yr^{-1}; they ranged from 9 to 2,386 eq ha^{-1}yr^{-1} (Figs. 4b and 6a). Critical loads for the best case scenario, calculated using the maximum weathering rates, were highest; 90 % were greater than 1,050 eq ha^{-1}yr^{-1}. Critical loads for the best case scenario ranged from 0 to 10,544 eq ha^{-1}yr^{-1} (Figs. 4c and 6a). The critical loads calculated using the mean weathering rates based on data from NSSC soil pits were consistently lower than for the other scenarios. For all states except New York, more than 80 % of plots fell into the range 200–780 eq ha^{-1}yr^{-1}; when New York was included, the upper end of the range increased to 1,150 eq ha^{-1}yr^{-1}. The mean critical loads ranged from 0 to 2,202 eq ha^{-1}yr^{-1} (Fig. 6a).

3.2 Exceedance (S+N)

For the mid-point weathering rate scenario, deposition exceeded the critical load in 45 % of plots (Table 3; Fig. 6b). For the worst case scenario, that value rose to 98 %, while for the best case scenario, deposition exceeded the critical load in only 15 % of plots. Exceedance of critical loads calculated using the mid-point weathering scenario mostly (>85 %) fell between −500 and 1,100 eq ha^{-1}yr^{-1}; the total range was −5,340 to 3,840 eq ha^{-1}yr^{-1} (Figs. 5a and 6b). Exceedance for the worst case scenario ranged from −1,186 to 4,889 eq ha^{-1}yr^{-1} (Figs. 5b and 6b). Exceedance for the best case scenario ranged from −9,343 to 3,026 eq ha^{-1}yr^{-1} (Figs. 5c and 6b). Exceedance for the NSSC mean scenario ranged from −657 to 1,760 eq ha^{-1}yr^{-1} (Fig. 6b). The percentage of plots in each state where the critical load was exceeded for the mid-point weathering scenario ranged from 13 to 62 %. For the worst case scenario, the percentage of plots by state where the critical load was exceeded ranged from 91 to 100 % (Table 2). For the best case scenario, the percentage of plots by state where the critical load was exceeded ranged from 1 to 27 % (Table 3). For the NSSC mean scenario, the percentage of plots by state where the critical load was exceeded ranged from 99 to 100 % (Table 3).

3.3 CL(N)$_{\text{nutrient}}$

Across the region, critical loads for nutrient N were low; over 90 % ranged from 200 to 300 eq ha^{-1}yr^{-1} (Fig. 7a). In Connecticut, over 80 % of critical loads ranged from 240 to 286 eq ha^{-1}yr^{-1}. In Massachusetts, 90 % of critical loads ranged from 210 to 306 eq ha^{-1} yr^{-1}. For Maine and New Hampshire, most critical loads (>80 %) ranged from 250 to 315 eq ha^{-1}yr^{-1}. New York had the lowest critical loads for nutrient N, ranging from 163 to 300 eq ha^{-1}yr^{-1} for 94 %. For Rhode Island, most critical loads were lower than 279 eq ha^{-1}yr^{-1}. The highest values were reported for Vermont, 20 % of the critical loads were greater than 325 eq ha^{-1}yr^{-1}.

Table 3 Percent of plots where the critical load (CL) is exceeded

	Percent exceedance				CL nutrient N
	CL S+N				
	Min	Mid	Max	Mean	
Connecticut	100	24	6	99	100
Massachusetts	100	56	9	99	99
Maine	91	15	3	99	99
New Hampshire	100	35	3	100	98
New York	100	62	27	100	100
Rhode Island	100	13	1	100	98
Vermont	99	56	20	99	94
New England and New York	98	45	15	>99	98

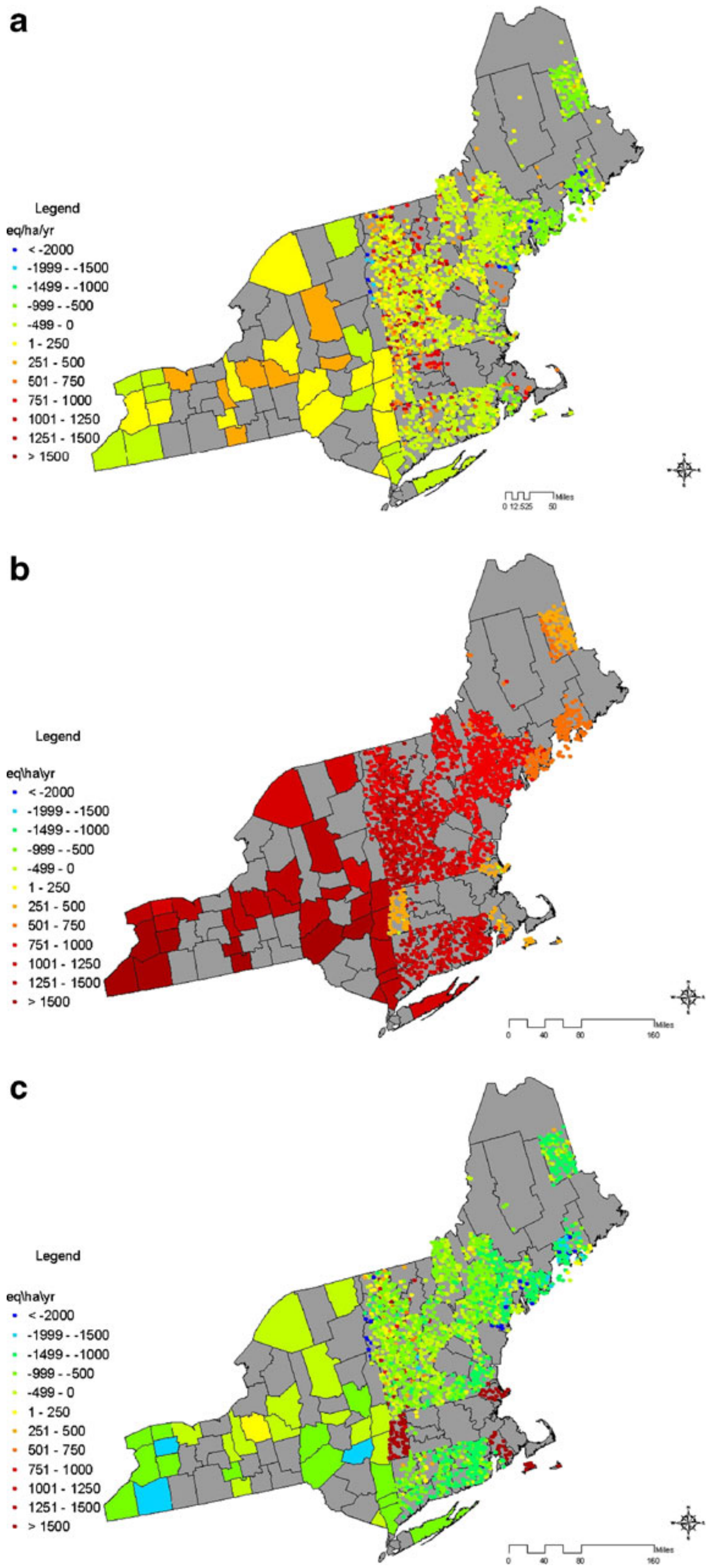

Fig. 5 Exceedance of critical loads for acidity (S+N) for sites in New England and New York. **a** Midpoint scenario. **b** Worst case scenario. **c** Best case scenario

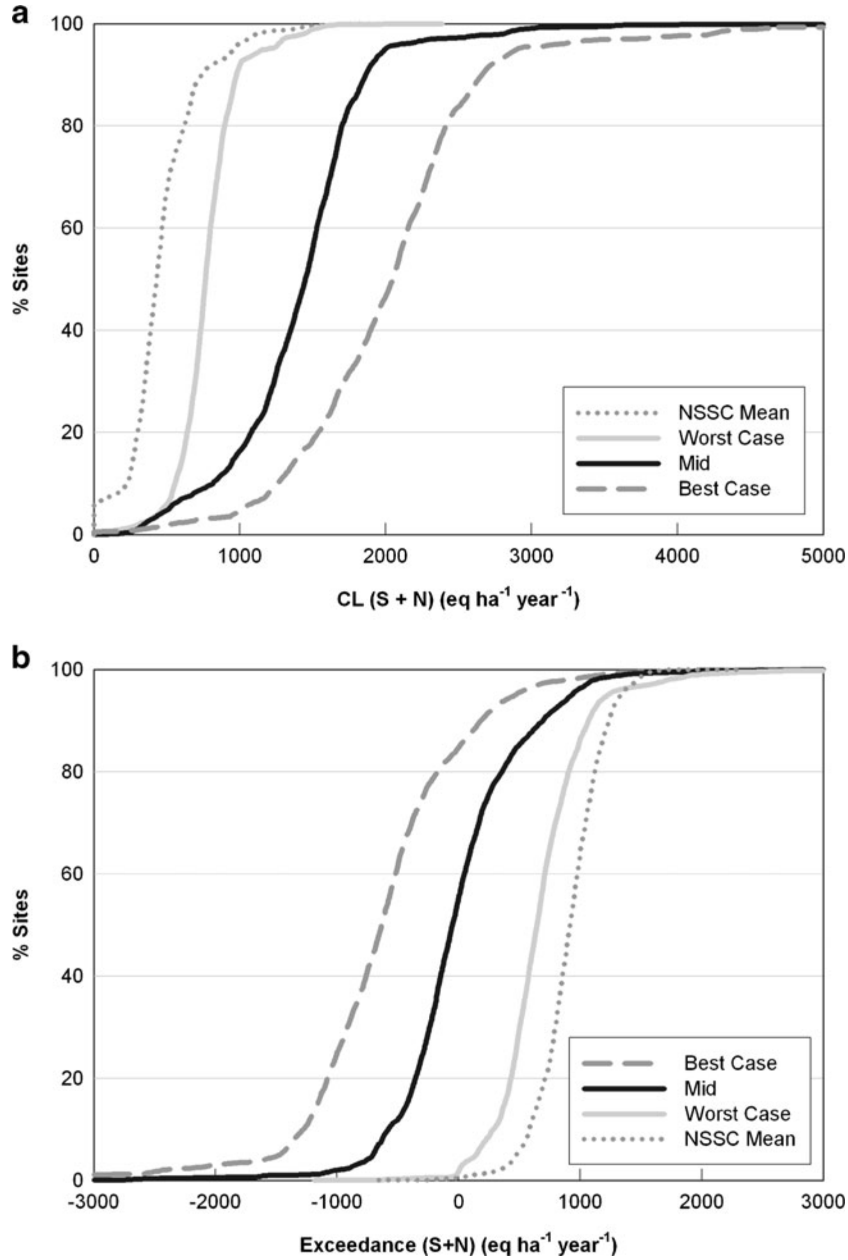

Fig. 6 Cumulative frequency of mid, worst case, best case, and NSSC mean (**a**) critical loads for acidity (S+N) and **b** exceedance of critical load for acidity for New England and New York. Calculated using midpoint, minimum, maximum, and NSSC mean soil mineral weathering rates. The critical load axis (**a**) does not display the full range, which extends to 10,544; neither does the exceedance axis (**b**) display the full range, which extends from −9,343 to 4,889

3.4 Exceedance $(N)_{\mathrm{nutrient}}$

Deposition exceeded critical loads for nutrient N for 98.5 % of the plots across the region; the percentage of plots by state where the critical load was exceeded ranged from 94 to 100 % (Fig. 7b; Table 3). Most exceedances across the region were less than 500 eq $ha^{-1} yr^{-1}$; they were lower for Maine, where deposition is lowest (Duarte et al. 2011a).

3.5 Forest Health

The objective of the forest health analysis was to determine whether the susceptibility of forest ecosystems to N and S deposition as quantified by critical load exceedance was related to measureable declines in forest health. We expected growth and crown density to be negatively correlated with exceedance; we expected declining vigor, crown dieback, and crown transparency

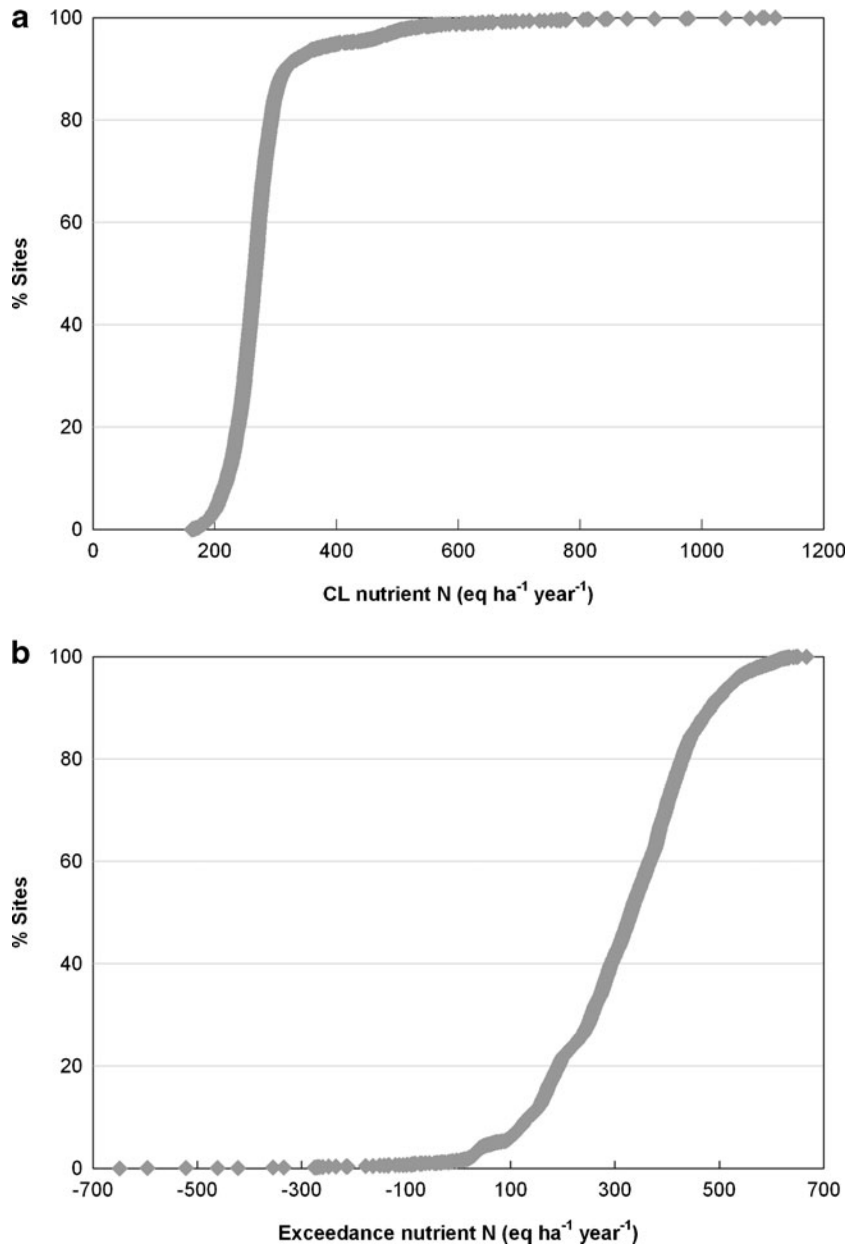

Fig. 7 Cumulative frequency of **a** critical loads for nutrient N and **b** exceedance of critical load for nutrient N for plots in New England and New York

to be positively correlated with exceedance. We observed significant negative correlations between critical load exceedance and growth for 17 species (Table 4). A slight negative trend for sugar maple (*Acer saccharum*) was not significant (n=3,310, p=0.06). Four species had positive correlations between growth and exceedance (Table 4). Quaking aspen (*Populous tremuloides*), red spruce (*Picea rubens*), and balsam fir (*Abies balsamea*) had the strongest correlations (>0.4) for crown dieback (the most reliable health indicator); these species also had high correlations with crown transparency (Table 5). Paper birch (*Betula papyrifera*) had a negative correlation with crown density and a positive correlation with crown dieback (Table 5). Northern red oak (*Quercus rubra*) had a positive correlation with crown transparency. Cherry (*Prunus*) and ash (*Fraxinus*) had positive correlations between exceedance and declining vigor. Several species exhibited correlations opposite to those expected (Table 5). Most species which showed damage by FH indicators, except quaking aspen, also showed decreased growth with exceedance.

Table 4 Growth versus exceedance by species for FIA (P2 plots) using Spearman's rank correlation analysis (α=0.05)

Species	Growth	*n*
American beech, *Fagus grandifolia*	−0.08	1,449
Balsam fir, *Abies balsamea*	−0.33	499
Bigtooth aspen, *Populus grandidentata*	−0.25	280
Black cherry, *Prunus serotina*	−0.07	1,026
Black oak, *Quercus velutina*	−0.18	257
Black spruce, *Picea mariana*	−0.44	41
Chestnut oak, *Quercus prinus*	−0.28	238
Eastern hemlock, *Tsuga canadensis*	−0.07	1,055
Eastern white pine, *Pinus strobus*	−0.19	1,464
Northern red oak, *Quercus rubra*	−0.11	1,533
Norway spruce, *Picea abies*	0.38	148
Paper birch, *Betula papyrifera*	−0.27	574
Pignut hickory, *Carya glabra*	0.44	31
Red maple, *Acer rubrum*	−0.11	3,861
Red spruce, *Picea rubens*	−0.09	616
Scarlet oak, *Quercus coccinea*	−0.26	170
Sweet birch, *Betula lenta*	−0.12	460
White ash, *Fraxinus americana*	−0.18	1,256
White spruce, *Picea glauca*	0.24	97
Yellow birch, *Betula alleghaniensis*	−0.12	669
Yellow poplar, *Liriodendron tulipifera*	0.35	47

3.6 Deposition Reduction

We considered three different deposition reduction scenarios: (1) 20 % reduction of both S and N deposition, (2) 50 % reduction in S and 26 % reduction in N, and (3) 48 % reduction in S and 32 % reduction in N. The percentage of sites in exceedance of the critical load for acidity (S+N) in the mid scenario decreased from 45 %, based on ClimCalc modeled deposition, to 23 % under the first scenario and to 13 % under the second and third scenarios. Exceedance of the critical load for acidity (S+N) for the NSSC mean weathering rate was not decreased substantially when deposition was reduced overall. In the worst case weathering rate scenario, the more stringent reduction scenarios (2 and 3) reduced exceedance from over 90 % to about 30 % in Maine and Rhode Island; regionally, the exceedance was reduced to 74–77 %. For nutrient N, deposition reductions did not significantly alter the proportion of sites were the critical load, $CL_{nut}N$, was exceeded, which included nearly all the sites in the region.

4 Discussion

Using different weathering scenarios to calculate critical loads led to a very broad range of critical loads. Similarly, for exceedance, there was little overlap between the extreme scenarios: in the worst case scenario, nearly all sites were exceeded (98 %), while for the best case scenario, only a relatively low proportion (15 %) of the sites were exceeded. The broad range spanned by the best case and worst case scenarios is not especially useful in a management or policy

Table 5 Vigor, crown density and dieback, and canopy transparency versus exceedance by species for FHM (P3 plots), HHS, NAMP, and VMC-FH using Spearman's rank correlation analysis (α=0.05)

	Vigor	*n*	Crown density	*n*	Crown dieback	*n*	Canopy transparency	*n*
American beech	0.13	433			0.12	502	0.30	502
Ash	0.22	189						
Balsam fir			−0.23	161	0.56	161	0.18	161
Cherry	0.33	47						
Eastern hemlock			0.36	82	−0.24	148		
Fir							−0.36	33
Northern red oak							0.27	208
Paper birch			−0.29	125	0.13	300		
Quaking aspen			−0.56	61	0.44	61	0.40	61
Red maple			−0.16	337	0.10	767	0.19	767
Red spruce			0.20	115	0.43	168	0.61	168
Sugar maple	0.06	3,408			−0.04	3,466	0.12	3,467
White ash	−0.32	49						

context. This is the central challenge posed by making critical load calculations at a broad scale: without extensive and complete soil and vegetation site measurements, one must rely on generalized maps which provide a range of the possible values of many parameters. While this range is very likely to include any value that might occur at a given site, it cannot predict the actual value well; this inaccuracy is exacerbated in heterogeneous landscapes. Thus, for assessing potential effects of atmospheric deposition for the whole region, the mid-point weathering scenario is the most valid because it is most likely to represent the typical condition of the region. However, the mid-point weathering scenario will not capture the most sensitive sites (e.g., those with the most acidic, shallow soils). In this section, we compare these critical loads estimates with other published values, we discuss factors that confound the patterns in critical loads, summarize the species patterns in forest health as a function of exceedance, and suggest ways in which critical load and exceedance estimates could be improved.

4.1 Comparisons to Other Critical Load Estimates

The critical loads for acidity (S+N) that we report for the mid scenario fell into similar ranges with other assessments. Critical loads for New England based on a detailed spatial analysis and modeling were reported in a similar range as this assessment, with the bulk of those values <3,000 eq $ha^{-1}yr^{-1}$ (NEG/ECP 2003; Miller 2005, 2006a, b, 2011). Critical loads were lower in southern New England and coastal areas (Miller 2005, 2006a, b). In a national assessment, the majority of the area in the northeastern USA had critical loads between 149 and 2,000 eq $ha^{-1}yr^{-1}$ (McNulty et al. 2007), although there were many more values in the 2,000–4,000 eq $ha^{-1}yr^{-1}$ range in their assessment. Ouimet et al. (2006) report a median critical load for Eastern Canada of 599 eq $ha^{-1}yr^{-1}$, with a reported range of 200 to >2,000 eq $ha^{-1}yr^{-1}$.

Our estimated critical load for nutrient N spanned a much narrower range of values and was much lower than the critical load for acidity, as has been reported in other assessments (Reinds et al. 2008). Empirical critical loads for N in the northeastern USA were from >215 to ~800 eq $ha^{-1}yr^{-1}$ for declines in growth and survivorship for some species, 570 eq $ha^{-1}yr^{-1}$ for increased nitrate leaching and <1,850 eq $ha^{-1}yr^{-1}$ for increased mortality and changes in species composition (Gilliam et al. 2011; Pardo et al. 2011a, b). One would expect the best correlation between the empirical critical load for the response of increased nitrate leaching from forests and the steady-state mass balance $CL_{nut}N$ because nitrate leaching is a component of the equation and can be a measure of N saturation. Nonetheless, the steady-state mass balance CL was generally at least 250 eq $ha^{-1}yr^{-1}$ lower than the empirical CL. This may suggest that the values we used for acceptable thresholds (for nitrate leaching and soil N immobilization) may not be valid for this region.

4.2 Sources of Variability in Critical Load Estimates

One challenge in estimating critical loads at the regional scale is that detailed data are not available on that scale; thus, it is necessary to estimate or model many of the key input parameters. We used the official soil series descriptions based on SSURGO maps because these were the most comprehensive data available to us at the time of the analysis. Because there is a large range between maximum and minimum values reported for each soil series, the resulting critical loads had an extremely large range.

The most significant example of this is for the mineral weathering rate, the most important parameter in determining the critical load for N+S (Whitfield et al. 2006; Hettelingh et al. 2007). Weathering, in this analysis, typically represents about 80 % of the critical load for acid inputs independent of N (called $CL(S)_{max}$; UBA 2004). Accurate determination of the mineral weathering rate is difficult because it requires specific information about the types and quantities of different minerals present (Hodson and Langan 1999). Given the spatial heterogeneity of soils in the northeastern USA (Lathrop et al. 1995), the weathering rate is highly variable and difficult to predict or model. There was considerably more variability in this parameter within each county than in other parameters. The range of weathering rates between the minimum and maximum scenarios can be quite large (in some cases more than 2,000 eq $ha^{-1}yr^{-1}$). The differences in the minimum and maximum potential clay percent can range from 5 % for a loamy sand-textured soil up to 40 % for a clay-textured soil. If a soil texture ranges from silty clay to clay, the difference in minimum and maximum clay percents can be as much as 60 %. Similarly, the differences in the minimum and maximum range for soil depth across the 326 soil types used in this analysis ranged from 15 to 152 cm with a mean difference of 50 cm. Because of these large ranges, we consider the mid scenario, based on the mid-point of the weathering rate range, to be most representative of the general patterns in the region.

We initially planned to use the NSSC soil pit mean as the main scenario since we assumed that using actual soil samples would constrain the ranges of the parameters so that the range observed would be considerably narrower than that between the best case and worst case scenarios. Instead, we found that all the NSSC soil pits clustered on the low end of the range for critical load or weathering based on soil series, often lower than the worst case scenario. This could be because the official soil series description is not representative of the soils that occur in New England and New York—that the soils in the northeastern USA are more sensitive than the soil series description indicates. Or, it may simply mean that the soil pit locations used by the NSSC are skewed towards sensitive sites and are not representative of the region. It is not possible to assess which explanation is correct based on the information that we have. However, given that the NSSC soil pits were randomly located and not sited to select for sensitive sites, one cannot ignore the possibility that the soils in the region may be considerably more sensitive than the official soil series suggests. Note that only FIA plots in areas with digitized soil maps available were included, which also makes the plots less representative than if all FIA had been included.

Other methodological issues may have affected the accuracy of the results. The most significant of these is the method and data used to estimate weathering. The clay percent-substrate method allows for separation of sites into weathering classes based on soil texture and mineral substrate (Sverdrup et al. 1990; Ouimet et al. 2006). In general, the clay percent method has been found to track weathering rates calculated using the PROFILE model when site data are used (Hodson and Langan 1999; Whitfield et al. 2006). The clay percent method was also used by Ouimet et al. (2006) in their analysis of critical loads for Eastern Canada as part of the NEG/ECP forest mapping group. Because our application of the clay percent method was based on soil series data rather than site-specific data, this represents a general approach; it cannot capture the spatial heterogeneity that exists across the landscape.

4.3 Forest Health Indicators

Twenty-one tree species in the northeastern USA exhibit detrimental impacts from atmospheric deposition as shown by the correlation between exceedance and forest health indicators (Tables 4 and 5, ESM Fig. 1). Some studies have identified relationships between growth or crown damage and exceedance (for example, Nelleman and Frogner 1994 and Ouimet et al. 2001), while others have found only weak or nonsignificant relationships (Augustin et al. 2005). Because the critical load equations are steady-state mass balances, they give no information about the timing of detrimental impacts. Thus, while sites with damage caused by atmospheric deposition should show exceedance, not all sites that have exceedance will necessarily exhibit the detrimental effects currently. The most likely mechanism for these impacts of atmospheric deposition is the sequence of ecosystem changes caused by acidification and cation depletion which lead to plant nutrient deficiencies and imbalances.

Plant nutrition regulates growth directly (Marschner 2002), but is also important in plants' ability to respond to environmental stresses. Several species that occur in the northeastern USA have well-documented mechanisms by which they are harmed as a result of the cation depletion that occurs with acidification. In many cases, nutrient deficiencies predispose trees to a secondary stress—which may vary with species. For red spruce, soil calcium depletion leads to reduced stress signaling capability and makes red spruce more susceptible to damage from winter injury (Halman et al. 2008; Hawley et al. 2006; Schaberg et al. 2002). The high correlation of crown dieback with exceedance for red spruce confirms that spruce on poorer sites are already being impacted by atmospheric deposition. Sugar maple is a commercially important species that grows across much of the region. At sites with low soil calcium, sugar maple has been shown to be susceptible to secondary stresses (Bailey et al. 2004, 2005; Bernier and Brazeau 1988a, b, c; Horsley et al. 2000); these stresses include pest defoliation (Horsley et al. 2000) and drought (Bauce and Allen 1991; Allen et al. 1992). Because of its commercial value for maple syrup, sugar maple is retained at suboptimal sites and managed more heavily than many species, which may make the patterns with exceedance more difficult to discern. Paper birch on sites with low soil calcium availability and high extractable soil aluminum in Vermont showed higher foliar loss, reduced fine branching of twigs, and increased tree mortality after the secondary stress of an ice storm (Halman et al. 2011).

Several nutrient-loving species, ash, cherry, sugar maple, and quaking aspen (Burns and Honkala 1990), as might be expected, showed more detrimental effects when exceedance was higher (at poorer sites); this was particularly evident for quaking aspen (Table 5). Quaking aspen survivorship was also reported to be negatively impacted by N deposition (Thomas et al. 2010) which may indicate that detrimental responses occur for both acidification and N saturation.

Improving northern red oak regeneration in the northeastern USA has been the object of considerable management effort (Buckley et al. 1998; Dey et al. 2008). These results suggest that northern red oak may not be viable on poor sites, a finding which is supported by an analysis in the Monogahela NF in WV, in which declines in basal areas from 1989–2000 were attributed to atmospheric deposition (Elias et al. 2009). A prior study had reported that low availability of Ca and K and high availability of Al combined with drought led to increased mortality and reduced growth of northern red oak in PA (Demchik and Sharpe 2000).

The high positive correlation for crown dieback and crown transparency and negative correlations for growth and crown density that we observed for balsam fir suggest that it may be at greater risk from atmospheric deposition than has been assumed. Balsam fir is cold tolerant, in contrast to red spruce, with which it often co-occurs (DeHayes et al. 1999). Thus, little research has focused on the susceptibility of balsam fir to atmospheric deposition. One study, however, in which chlorophyll fluorescence was measured, indicates that balsam fir is stressed by low soil Ca availability (Boyce 2007). Similarly, Van Doorn et al. (2011) report high mortality of balsam fir in an assessment across the Hubbard Brook Valley, NH. The secondary stress, mechanism, or condition that has led to this decline is unknown and merits further investigation.

Attributing causes for forest decline remains complex because many factors at local, regional, and global scale may influence forest health. For example, Van Doorn et al. (2011) observed a significant decrease in growth of yellow birch across the Hubbard Brook Valley which they attribute to secondary succession. The negative correlations for yellow birch that we observed between growth and exceedance (Table 4) and that Thomas et al. (2010) report between survivorship and N deposition, however, suggest that atmospheric deposition is implicated, to some extent, in this decline. While most of the significant growth response had a negative correlation with exceedance, four species had positive correlations. One possible explanation for the positive correlation of growth with exceedance is that these species may be benefitting from the declines of the species with which they co-occur. Certainly, climate change and N deposition have been shown to increase growth of some tree species (Dietze and Moorcroft 2011; Thomas et al. 2010; Zhang et al. 2012) across the northeastern USA.

4.4 Improving Critical Loads and Exceedance Estimates

The biggest gains in accuracy of critical loads could be made by increasing the accuracy of weathering rate estimates. Several steps would facilitate this. A first step would be integrating detailed soil sampling and chemical/mineralogical analysis into a national assessment program such as FIA. Soil samples would need to be collected by horizon and to bedrock (or at least rooting depth). In Sweden, which has an extensive network of soil pits with mineralogy information (>60,000 sites), estimates of mineral weathering rate can be made with certainty (Akselsson et al. 2004). A second step would be to improve soil maps so that the ranges of parameters would be more constrained within smaller landscape units. A third step would be to improve the calculation of weathering by using a dynamic model such as PROFILE (Sverdrup and Warfvinge 1993; Warfvinge and Sverdrup 1992) coupled either with data from actual sites or more constrained soil type data. Finally, a significant improvement would be to get accurate data on soil depth, for example an accurate map of soil depth could increase the accuracy of the weathering rate tremendously.

For estimating exceedance, the main limitation (beyond the accuracy of the critical load estimates) is the accuracy of deposition measurements. The deposition model that we used in this analysis, ClimCalc (Ollinger et al. 1993), will not capture the highest deposition values because cloud and fog deposition are not included and the impact of elevation and tree species on deposition are not modeled. For example, a model which includes these factors, the High-Resolution Deposition Model (Miller et al. 1993; Miller 2000), predicts hotspots in N deposition, in southern Vermont, for example, of >30 kg ha^{-1}

year^{-1}, while ClimCalc predicts <12 kg ha^{-1}year^{-1} (Ollinger et al. 1993), and NADP maps wet-only deposition in the region as 4–6 kg ha^{-1}year^{-1} (NADP 2009). Clearly, this range of deposition values would have a dramatic effect on the level of exceedance reported.

Another way that this process could be improved would be to refine the critical thresholds used in calculating the critical load, especially those used for the acceptable ANC leaching term (Aherne et al. 2001; Hall et al. 2001; UBA 2004; Reinds et al. 2008). Refining the values used for the BC/Al threshold for different forest types based on physiological responses observed would strengthen the link between critical load and ecosystem response. Similarly, as has been done in the Netherlands (de Vries et al. 2007), evaluating the relationship between soil solution nitrate concentration and detrimental physiological responses in different ecosystem types would help refine the acceptable N leaching term and, thus, improve estimates of the nutrient N critical load.

5 Conclusions

In trying to estimate critical loads for a broad region, there are inevitable tradeoffs between trying to cover a large area (with few site-specific data) and accurately representing a specific location. This assessment does a better job of capturing the typical values than in identifying the most susceptible ecosystems. Thus, caution should be taken in interpreting values to give specific information at a point in space. Improving regional soil datasets would most improve critical loads and exceedance estimates. Nonetheless, the strong relationships between forest health indicators and exceedance indicate that atmospheric deposition continues to detrimentally impact forest health in the northeastern USA. The species most affected are balsam fir, red spruce, quaking aspen, and paper birch.

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