Empirical Critical Loads of Atmospheric Nitrogen Deposition for Nutrient Enrichment and Acidification of Sensitive US Lakes

JILL S. BARON, CHARLES T. DRISCOLL, JOHN L. STODDARD, AND ERIC E. RICHER

The ecological effects of elevated atmospheric nitrogen (N) deposition on high-elevation lakes of the western and northeastern United States include nutrient enrichment and acidification. The nutrient enrichment critical load for western lakes ranged from 1.0 to 3.0 kilograms (kg) of N per hectare (ha) per year, reflecting the nearly nonexistent watershed vegetation in complex, snowmelt-dominated terrain. The nutrient enrichment critical load for northeastern lakes ranged from 3.5 to 6.0 kg N per ha per year. The N acidification critical loads associated with episodic N pulses in waters with low values of acid neutralizing capacity were 4.0 kg N per ha per year (western) and 8.0 kg N per ha per year (northeastern). The empirical critical loads for N-caused acidification were difficult to determine because of a lack of observations in the West, and high sulfur deposition in the East. For both nutrient enrichment and acidification, the N critical load was a function of how atmospheric N deposition was determined.

Keywords: nitrogen, critical load, lakes, nutrient enrichment, acidification

Atmospheric deposition of nitrogen (N) from fossil-fuel and agricultural emissions is the largest single source of N in the northeastern United States and in all US mountain regions (Fenn et al. 2003, Howarth 2008). The environmental risks associated with elevated atmospheric N deposition have long been recognized in Europe, where N deposition critical loads have been used as a guidance tool for air quality management since the 1990s (Hettelingh et al. 2009). The US Environmental Protection Agency (EPA) is currently in the process of developing a framework to evaluate the exposure of ecological receptors to nitrogen oxides (NO_x) and other pollutants and to guide the recovery of affected ecosystems, drawing on the large body of literature that has amassed over the past several decades (USEPA 2008, 2009, Pardo et al. 2011). The conclusion reached from these assessments of ecological effects is that current air quality standards are inadequate to protect ecosystems from the effects of atmospheric N deposition. It is timely, then, to summarize the knowledge of N deposition effects on freshwater lake ecosystems and to propose *critical loads*, or thresholds below which there are no discernible effects (Burns et al. 2008).

We present critical loads for N deposition for headwater lakes in regions of the United States where the primary environmental impacts come from N deposition and climate change as opposed to land use or water pollution discharges. Nitrate (NO_3^-) concentrations in otherwise undisturbed lakes are often related to atmospheric deposition in the absence of N-fixing riparian vegetation; there is a strong positive correlation between lake NO_3^- concentrations and the amount of atmospheric N deposition (Bergström and Janssen 2006, Elser et al. 2009). We empirically derived N critical loads from measured chemical and biological changes in N deposition using published literature and data sets. *Reactive N*, or that which can be readily metabolized by microbes and algae, follows complex cycles through the atmosphere and ecosystems (Galloway et al. 2003) and has two distinct ecological effects in lakes: nutrient enrichment and acidification. We based our selection of N critical loads on the following logical sequence: (a) In otherwise undisturbed headwater lakes with low N deposition, late-summer lake NO_3^- is also low in concentration. (b) Increasing amounts of N deposition in otherwise undisturbed headwater lakes are positively correlated with increased late-summer lake NO_3^- concentrations. (c) Increased lake NO_3^- concentrations, or nutrient enrichment, triggers biological responses, including increased algal productivity and loss of diversity. Finally, (d) increased lake NO_3^- concentrations can also represent the loss of watershed acid-neutralizing capacity (ANC), and this triggers lake acidification and a loss of diversity. Whether lakes first exhibit symptoms of nutrient enrichment and then acidify or display characteristics of only acidification is a function of the starting, or antecedent, conditions of the ecosystem (figure 1).

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Nitrogen limitation of algal productivity is or was widespread in undisturbed freshwaters of the Northern Hemisphere (Elser et al. 1990, Bergström and Jansson 2006). Lake ecosystems that respond to nutrient enrichment have very low initial N concentrations. Productivity increases somewhat when N becomes available but is truncated as phosphorus (P) becomes the limiting factor for growth (Elser et al. 2009). However, even slight increases of N in environments that evolved with strong N limitation stimulate plant growth and can decrease species richness by giving some plants and algae a competitive advantage over others (Bobbink et al. 2010). Consequences include greater primary productivity, altered algal assemblages, and less algal diversity, with possible implications for food webs because P-limited algae make poor-quality food for herbivorous zooplankton (Nydick et al. 2004, Elser et al. 2009). Lake nutrient enrichment proceeds in forested catchments after terrestrial N demand is met and saturation has occurred (figures 1 and 2a; Stoddard 1994, Aber et al. 2003). However, in some high-elevation lakes in the West with limited or unvegetated catchment areas, N enrichment may be an immediate direct consequence of even slight increases in atmospheric deposition (figures 1 and 2b; Fenn et al. 2003, Elser et al. 2009).

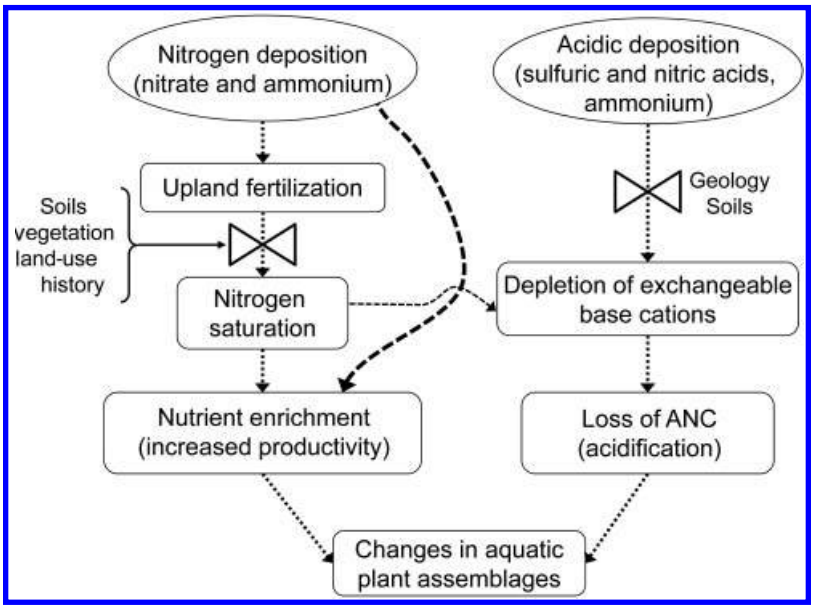

Figure 1. Conceptual model of the influence of atmospheric nitrogen (N) deposition and acidic deposition on freshwaters. In western lakes with little upstream vegetation, the responses to N deposition can be rapid. In eastern lakes, responses can take much longer, possibly decades or centuries. Abbreviation: ANC, acid-neutralizing capacity.

Acidification occurs when deposition of either acids (primarily sulfuric and nitric acids) or N as NO_3^- or ammonium (NH_4^+) exceeds the ability of watershed soils, vegetation, and surface waters to neutralize or retain these inputs (figure 1). Both acidic deposition and N saturation acidify freshwaters (Reuss and Johnson 1986, Charles and Christie 1991, Stoddard 1994). Catchment sensitivity to acidic deposition is largely governed by bedrock and surficial geology and soils, and much of the United States is insensitive to acidic deposition (Charles and Christie 1991). The regions of most concern for surface-water acidification by acidic deposition are areas underlain by intrusive igneous rocks, typical of the western mountains, the upper Midwest, and parts of the Northeast, or base-poor sandstones typical of the Northeast and the mid-Atlantic and the central and southern Appalachian mountains (Charles and Christie 1991). High-elevation catchments with shallow soils often exhibit limited acid neutralization associated with the supply of base cations. Catchment sensitivity is also influenced by

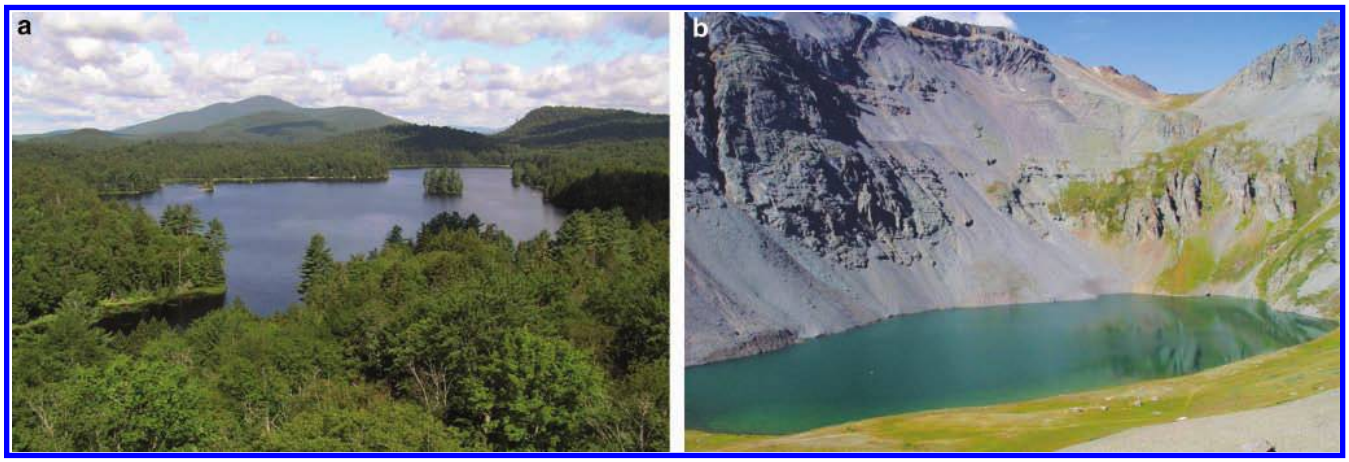

Figure 2. (a) Arbutus Lake, in the Adirondacks, is typical of an eastern lake with a forested catchment. Photograph: Charles T. Driscoll. (b) Clear Lake in the Colorado Rocky Mountains is an alpine lake with little upstream vegetation. Photograph: James J. Elser.

past acidic deposition or land use that diminishes the ability of soils to neutralize ongoing atmospheric deposition because of long-term declines in exchangeable base cations or accumulation of sulfate (Driscoll et al. 2001).

Mechanisms and evidence for N deposition–induced nutrient enrichment

A survey of the literature shows that nutrient-poor waters are commonly N limited, especially undisturbed northern temperate, boreal, or high-altitude lakes that receive low atmospheric N deposition (Elser et al. 1990, Bergström and Jansson 2006, Elser et al. 2009). A meta-analysis of lakes from 42 regions of Europe and North America showed a consistent pattern: N limitation for atmospheric deposition below approximately 2.5 kilograms (kg) N per hectare (ha) per year, colimitation of N and P for deposition between about 2.5 and 5.0 kg N per ha per year, and P limitation in areas with N deposition greater than 5.0 kg N per ha per year (Bergström and Jansson 2006). Similar results were found by exploring the N:P resource response ratios among lakes receiving greater or lesser amounts of N deposition (Elser et al. 2009). Bergström and Jansson (2006) and Elser and colleagues (2009) suggested that the majority of lakes in the Northern Hemisphere may have originally been N limited and that atmospheric N deposition has changed the balance of N and P in lakes such that P limitation is commonly observed today.

Data from the eastern (Linthurst et al. 1986) and western (Eilers et al. 1987) lake surveys were evaluated to determine the extent of N limitation caused by atmospheric deposition in the United States (table 1). These surveys were conducted on a subset of US lakes with low acid-neutralizing capacity in order to evaluate the current and potential future extent of damage from acidic deposition. Although the surveys are several decades old, they remain the most extensive and uniformly collected lake data available for a national assessment. In these surveys, samples were collected during late summer and autumn to minimize the interference of snowmelt or watershed nutrient-cycling processes in interpreting the atmospheric deposition–lake chemistry relationship.

Lake management guidelines often recommend the use of total N (TN) and total P (TP) concentrations and their ratio to indicate the lake's trophic state and the degree of limitation by a single nutrient (Dodds 2003). Evaluating the extent to which atmospheric N deposition influences lake nutrient dynamics, however, is a different question—one directly related to air quality. In a recent article in which TN:TP was compared with the ratio of dissolved inorganic N (DIN; the sum of NO_3^- and NH_4^+) to TP in lakes specifically receiving N deposition, Bergström (2010) found that the DIN:TP ratio varied specifically with inorganic N deposition. The more commonly used TN:TP ratio was less sensitive to changes in N deposition because it was additionally responsive to catchment processes that retain or release organic and inorganic N to lakes. For this reason, we used the DIN:TP ratio to evaluate the proportion of otherwise undisturbed lakes affected by atmospheric N deposition. There were N-limited lakes—defined by Bergström (2010) as DIN:TP $\leq$ 1.5 by mass—in every region of the country, but the greatest proportion of N limitation occurred in the Rockies (25%), the Sierra Nevada and Cascades (22%), and the upper Midwest (22%; table 1). These lakes, by definition, are susceptible to chemical and biological change from atmospheric N deposition.

Table 1. Proportion of lakes in three nutrient limitation classes (N limited, P limited, and intermediate) based on ratios of dissolved inorganic nitrogen (DIN; nitrate + ammonium) to total phosphorus (TP).

		N-limited lakes[b]		P-limited lakes[c]		Lakes with intermediate N:P ratios[d]	
Region	Total number of lakes[a]	Number	Percentage	Number	Percentage	Number	Percentage
Adirondacks	1290	41	3	1082	84	137	13
New England	4361	330	8	2891	66	1139	26
Poconos/Catskills	1506	105	7	899	60	502	33
Southeast	2424	280	11	1689	70	455	19
Upper Midwest	8755	1890	22	5433	63	1251	15
Rockies	6666	1641	25	3668	55	1357	20
Sierra/Cascades	4155	934	22	2687	65	534	13
Total	28,976	5220	18	18,350	63	5407	19

Source: The data are from the Eastern Lake Survey (Linthurst et al. 1986), conducted in the fall of 1984, and the Western Lake Survey (Eilers et al. 1987), conducted in the fall of 1985.

[a]The Eastern and Western Lake Surveys were stratified random samples of all lakes less than 4 hectares in size; the estimates of the number of lakes in each region are based on the target population sizes for each survey.

[b]Lakes with DIN:TP ratios (by weight) less than 1.5 were characterized as N limited, on the basis of the work of Bergström (2010).

[c]Lakes with DIN:TP ratios (by weight) greater than 4 were characterized as P limited, on the basis of the work of Bergström (2010).

[d]Lakes with DIN:TP ratios (by weight) between 1.5 and 4 could not be assigned to a nutrient-limitation class and are characterized as either colimited or limited by something other than N or P.

N, nitrogen; P, phosphorus

Phytoplankton are the aquatic biota most likely to respond rapidly to N deposition in lakes that historically received low nutrient inputs, because they are primary producers with life cycles of days to weeks. Increases in phytoplankton biomass in lakes with increasing N deposition or experimental N additions have been reported in a number of undisturbed mountain or forested catchments in the United States and Europe (Sickman et al. 2003, Nydick et al. 2004, Bergström and Jansson 2006, Elser et al. 2009). Paleolimnological studies in the western United States have shown increased productivity in lakes over time (using cell counts and pigments as proxies) commensurate with increased N deposition; no productivity responses have been noted where there is no indication of a trend in N deposition (Wolfe et al. 2003, Das et al. 2005).

The loss of biodiversity, defined as a decrease in overall species richness and evenness, in lake algae commensurate with elevated N deposition has been reported in a growing number of studies, similar to results related to terrestrial vegetation (Bobbink et al. 2010). Interlandi and Kilham (1998) demonstrated that maximum species diversity was maintained when N concentrations were below 0.7 micromoles (μmol) of NO_3^- per liter (L) in lakes in the Yellowstone National Park (Wyoming, Montana) region. Marked community shifts in phytoplankton have been observed in western lakes in response to N deposition and to short-term N enrichment experiments at concentrations of from 1.4 to 17.2 μmol NO_3^- per L (Pardo et al. 2011). Results from recent studies include a change from chrysophyte phytoplankton in low-N lakes to cyanophytes and chlorophytes in lakes with higher N and an increased representation of mesotrophic diatom species after 1950 (Jassby et al. 1994, Wolfe et al. 2003, Lafrancois et al. 2004, Saros et al. 2010). Two species of diatom, *Asterionella formosa* and *Fragilaria crotonensis*, now dominate the flora of several alpine and montane Rocky Mountain and Sierra Nevada lakes where there has been an increase in N deposition (Interlandi and Kilham 1998, Wolfe et al. 2003, Saros et al. 2010). By hindcasting wet N deposition between 1950 and 1960, Baron (2006) proposed that 1.5 kg N per ha was sufficient to alter diatom assemblages from the original composition to a new assemblage dominated by *A. formosa* and *F. crotonensis*; this value was recently independently derived through similar methods applied to lakes in the Sierra Nevada and in Yellowstone National Park (Saros et al. 2010). The growth of *A. formosa* and *F. crotonensis* has been stimulated with N amendments during *in situ* incubations, using bioassays and mesocosms, whereas other species have shown negative growth responses to increased N (McKnight et al. 1991, Wolfe et al. 2003, Lafrancois et al. 2004, Saros et al. 2005, Michel et al. 2006).

Mechanisms and evidence for N-induced acidification

Surface-water sensitivity to acidification is commonly measured in terms of pH and ANC. Both of these indicators decrease as lakes acidify, and waters are classified as acidic when ANC values become negative. Acidification can be further characterized as *chronic acidification* (when lakes are acidic year-round; ANC < 0 microequivalents [μeq] per L) or *episodic acidification* (short-term decreases in pH and ANC, lasting on the order of hours to weeks). In water bodies with low ANC, strong hydrologic events that temporarily decrease ANC cause susceptibility to acid episodes. Although N deposition can influence both chronic and episodic acidification, its effects are most marked on episodic acidification (Schaefer et al. 1990, Murdoch and Stoddard 1992, Wigington et al. 1996).

Episodic acidification from atmospheric N deposition has mostly been reported from snow-dominated environments where winter N deposition accumulates in the snowpack. Episodic acidification associated with NO_3^- leaching can also occur from summer and fall rain events (Wigington et al. 1996). Snowmelt or rain events trigger pulses of reactive N to streams and lakes (Stoddard 1994). Sources of NO_3^- pulses to streams and lakes include preferential elution of solutes including N from the snowpack, leaching of N-rich soil solutions from mineralization, and nitrification.

Severe episodes can produce conditions that are as deleterious to biota as chronic acidification and can be important in acid-sensitive regions (Baker et al. 1996). Roughly 10% of Adirondack lakes were chronically acidified in the early 1990s, but more than three times that number (31%) exhibited acid episodes (Driscoll et al. 2001). Pulses of NO_3^- were ranked as the second most important cause of short-term acidification in the Adirondack and Catskill Mountains; only dilution of base cations, which occurs in virtually all surface waters under increased flow, ranked higher (Wigington et al. 1996). There are no chronically acidic lakes in the western United States, but acid episodes have been observed in both the Colorado Front Range and the Sierra Nevada, where NO_3^- leaching during snowmelt decreases ANC to 0 μeq per L or below (Stoddard 1995, Williams and Tonnessen 2000).

Critical loads for chemical responses to N deposition

Empirical cross-site watershed studies show the relationship between lake N concentrations or N export as a function of atmospheric N deposition (Sickman et al. 2002, Aber et al. 2003, Fenn et al. 2003, Bergström and Jansson 2006). Such cross-site analyses were originally conducted for Europe (Wright et al. 2001) and typically show a "dog-leg" pattern—low surface-water NO_3^- concentrations or leaching with low levels of N deposition and elevated leaching losses and concentrations above a deposition threshold. For European surface waters, that threshold was 10 kg N per ha (Wright et al. 2001). Keep in mind that the threshold will differ between lakes of strongly and those of weakly vegetated catchments, such as those shown in figure 2; the inflection point where NO_3^- concentrations increase in response to increasing N deposition defines the empirical N critical load.

Lake N concentrations. We obtained lake NO_3^- concentrations from three regions of the United States where lakes have been protected from land-use change in parks, national forests, or wilderness. The northeastern United States, the

Rocky Mountains, and the Sierra Nevada are lake-rich regions where atmospheric deposition is the dominant—if not the sole—source of inorganic N. Lake chemistry data were obtained from several sources. The Northeast lake data (n = 216) were from Aber and colleagues (2003). These data represent lakes in forested catchments of Vermont, New Hampshire, Maine, and the Adirondack and Catskill Mountains of New York. The Rocky Mountain (n = 286) and the Sierra Nevada (n = 20) lakes were selected from the US Forest Service's Air Resource Management online database (*www.fs.fed.us/ARMdata*). The Sierra Nevada lake set was augmented with 10 additional lakes from Sickman and colleagues (2002), for a total of 30 lakes. Western lakes were included if they were located above the tree line (alpine) and had recorded NO_3^- concentrations from August, September, and October for the years 1997–2006. The mean 1997–2006 NO_3^- concentration for each lake (table 2; also see the see the supplementary online materials at *dx.doi.org/10.1525/bio.2011.61.8.6*) was related to N deposition amounts in the section below. The mean lake concentrations of 14.4 (standard deviation [SD] = 0.8), 3.7 (SD = 1.9), and 2.7 (SD = 1.1) µmol N per L for the Northeast, Rocky Mountain, and Sierra Nevada lakes, respectively, were lower than the predicted median total N (NO_3^- plus NH_4^+ plus organic N) concentration of 17.9 µmol N per L for streams and rivers of the Northeast and were much lower than the median total N value of 13.6 µmol N per L for western mountain streams and rivers reported by Smith and colleagues (2003). The values from Smith and colleagues (2003) include N from atmospheric deposition. When these investigators modeled background N concentrations in the absence of human disturbance for headwater streams that correspond to our headwater lakes, the predicted background N concentrations for waters from all three of our lake regions ranged 0 to 2.4 µmol N per L. The upper value is similar to our measured values from the Sierra Nevada, slightly lower than the Rocky Mountain mean lake concentration, and much lower than the Northeast mean lake concentration. Smith and colleagues (2003) interpreted the deviation from the modeled background concentrations as evidence of cultural N loading, including atmospheric deposition.

How much N deposition is there? We explored the relationship between atmospheric N deposition and the average NO_3^- concentrations for the Northeast, Rocky Mountain, and Sierra Nevada lakes by first confronting the issue of how best to quantify atmospheric N deposition. The National Atmospheric Deposition Program (NADP; 2010) is the most commonly used metric of atmospheric deposition. NADP reported wet-only concentrations and deposition amounts for inorganic N at 186 individual sites across the United States. Few of these sites are collocated with the lakes of interest. Wet inorganic N deposition is calculated for each site by multiplying concentration by site-specific precipitation. National-scale deposition maps are produced by a linear interpolation between sites. A drawback of this approach is its inability to capture orographic precipitation in mountainous regions, and this can be seen in the smooth transitions of mean annual 1997–2006 wet N deposition, especially in the Sierra Nevada, Rocky Mountains, and Northeast (identified with ovals from west to east, respectively) in figure 3a. Summary statistics can be found in the supplementary materials.

Greater spatial resolution of wet inorganic N deposition was produced by combining the N from NO_3^- and NH_4^- concentrations from NADP with PRISM (Parameter-elevation Regressions on Independent Slopes Model) precipitation values (hereafter, PRISM+NADP) for 1997–2006 (figure 3b and supplementary materials). PRISM provides continuous, high-resolution digital grid estimates of climatic parameters that resolve orographic precipitation amounts in complex terrain (*www.prism.oregonstate.edu*). Because annual PRISM data sets for precipitation have a

Table 2. Summary of mean lake nitrate (NO_3^-) concentrations (in micromoles per liter), inorganic nitrogen (N) deposition amounts (in kilograms [kg] of N per hectare [ha] per year), and nutrient enrichment inflection points (in kg N per ha per year) where lake NO_3^- concentrations reflect increased N deposition and episodic acidification inflection points.

Region	Mean lake NO_3^-	Mean 1997–2006 NADP N deposition	Nutrient enrichment inflection point (NADP)	Mean 1997–2006 (PRISM+NADP) deposition	Nutrient enrichment inflection point (PRISM+NADP)	2002 total N deposition	Nutrient enrichment inflection point (total N)	Episodic acidification inflection point
Sierra Nevada (n = 30)	2.7 (1.10)	1.5 (0.22)	1.5	2.5 (0.34)	1.5	3.4 (0.47)	2.0	—
Rocky Mountains (n = 285)	3.7 (1.90)	1.2 (0.27)	1.0	3.0 (0.28)	2.0	4.1 (0.24)	3.0	4.0
Northeast (n = 216)	14.4 (0.76)	4.7 (0.18)	3.5	5.2 (0.18)	3.5	9.8 (0.20)	6.0	8.0

Source: Aber et al. 2003, Sickman et al. 2002, US Forest Service's Air Resource Management online database (*www.fs.fed.us/ARMdata*).
Note: Inorganic N deposition was calculated three ways, as was described in text and as is shown in figure 3. Coefficients of variation are shown in parentheses.
NADP, National Atmospheric Deposition Program; PRISM, Parameter-elevation Regressions on Independent Slopes Model; PRISM+NADP, concentrations from NADP with PRISM precipitation values

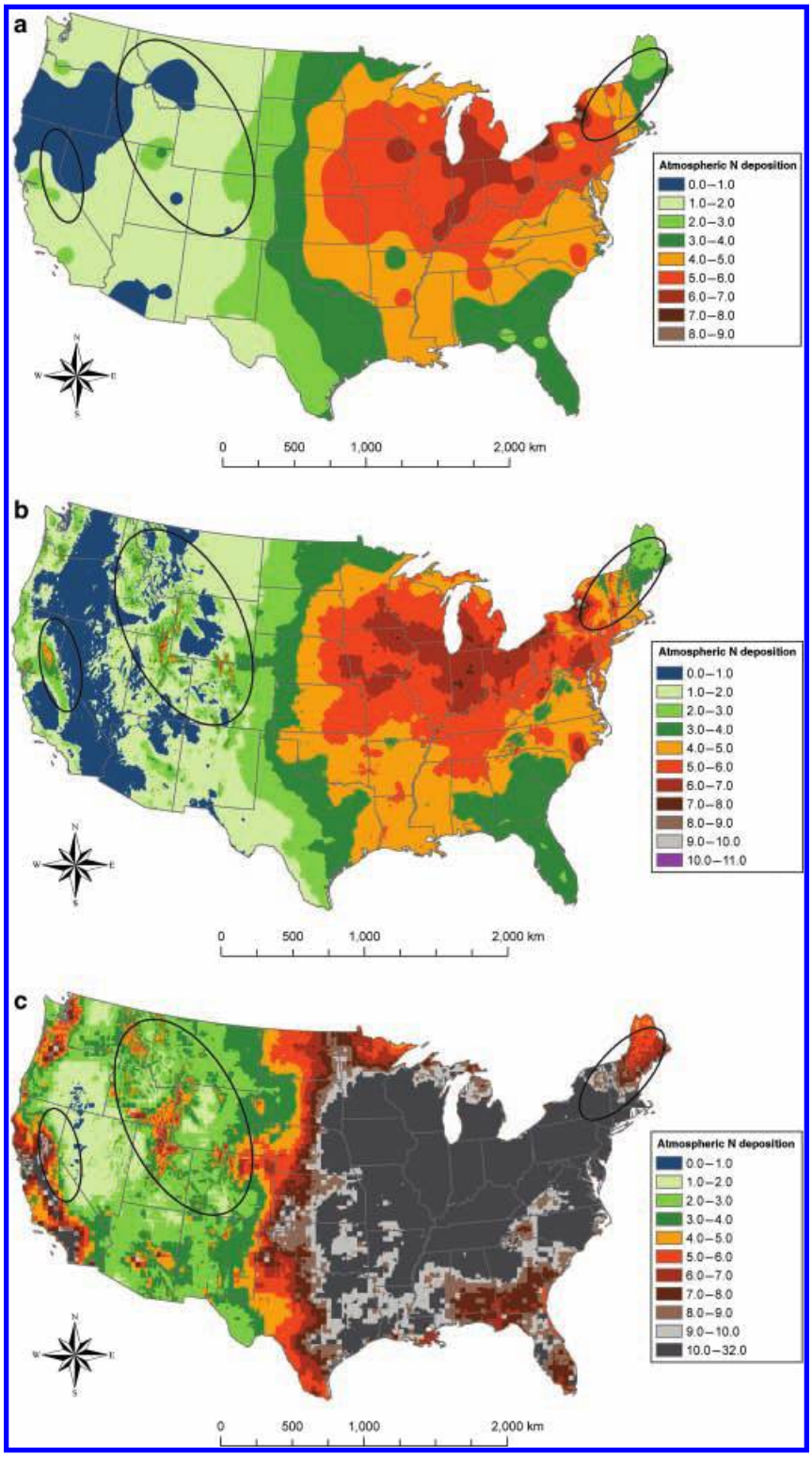

Figure 3. Three ways of presenting atmospheric nitrogen (N) deposition in kilograms per hectare per year for the conterminous United States. (a) 1997–2006 average wet N deposition from the National Atmospheric Deposition Program (NADP; 2010). (b) Deposition calculated from the 1997–2006 mean PRISM (Parameter-elevation Regressions on Independent Slopes Model) precipitation and NADP wet N (NO_3^-–N and NH_4^+–N) concentrations (PRISM+NADP); and (c) PRISM+NADP wet plus Community Multiscale Air Quality (CMAQ) dry (total N) deposition for 2002 only. Abbreviation: km, kilometers.

grid resolution of approximately 4 kilometers (km), whereas NADP isopleth maps for annual N concentration have a grid resolution of 2.5 km, the PRISM data sets were resampled to the NADP grid resolution of 2.5 km. The average wet inorganic N deposition in 2002 derived from PRISM+NADP compares well with the wet deposition values observed at the 186 NADP sites that met NADP data completeness criteria in 2002 (figure 4a). This pattern suggests that using PRISM precipitation data sets to derive deposition values for topographically complex areas such as the Rocky Mountains and the Sierra Nevada provides an appropriate representation of N deposition to mountain regions, which is certainly a more logical approach than using deposition data that are not topographically resolved.

Improved representation of wet inorganic N deposition is still wet-only deposition. Many regions of the United States receive equal or greater amounts of N deposition from dry deposits (Fenn et al. 2003, Weathers et al. 2006). An estimate of total inorganic N deposition can be derived only from the summation of wet and dry deposition. We considered including dry deposition measurements from the Clean Air Status and Trends Network (CASTNET; *www.epa.gov/castnet*), for which data are available for the years 1997–2006. We rejected combining CASTNET data with the wet N deposition data because there are only 86 sites nationwide in the CASTNET data set, and although gridded CASTNET concentrations for several gaseous species are available, gaseous ammonia (NH_3)—a large component of N deposition—was not measured. Instead, we extracted the dry deposition component from the Community Multiscale Air Quality (CMAQ; *www.cmaq-model.org*) model, which produces total (wet plus dry) N deposition values for the conterminous US, and combined the CMAQ dry value with the PRISM+NADP wet value to estimate total inorganic N deposition (figure 3c and the supplementary materials). CMAQ wet-plus-dry model output was considered unacceptable for our purposes because of the coarse 36-km spatial resolution. CMAQ data

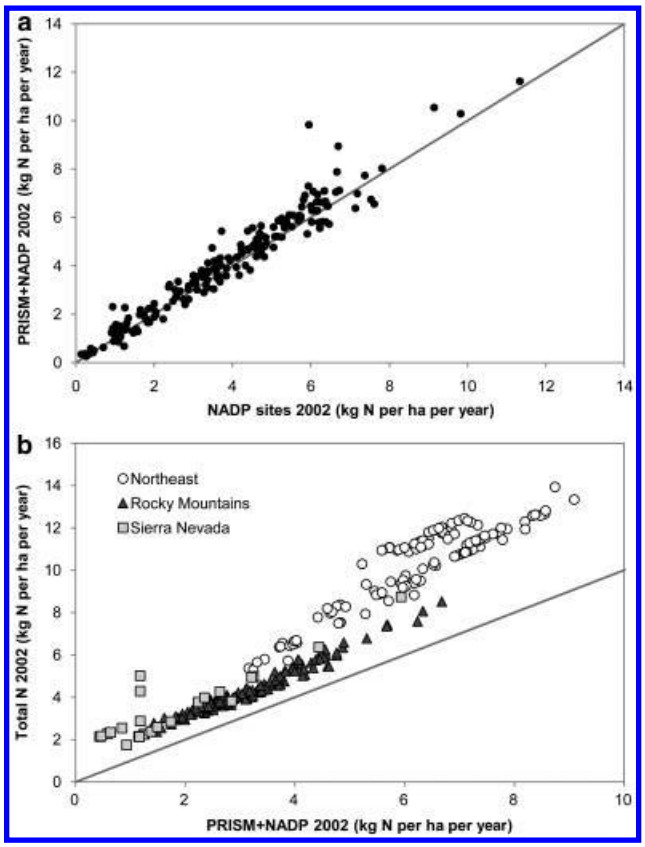

Figure 4. Comparison of nitrogen (N) deposition estimates, in kilograms (kg) N per hectare (ha) per year. (a) 2002 mean wet inorganic N deposition from National Atmospheric Deposition Program sites (NADP; 2010) with 2002 mean PRISM (Parameter-elevation Regressions on Independent Slopes Model) precipitation and NADP wet N concentrations (PRISM+NADP). (b) 2002 PRISM+NADP with 2002 total N.

are only available for the year 2002. A comparison of the PRISM+NADP wet and CMAQ dry (hereafter, total N) deposition with PRISM+NADP at the locations of the lakes in our analysis for 2002 showed that wet-only deposition underestimated deposition for all regions, but the underestimation was most pronounced for lakes in the Northeast (figure 4b). The inclusion of dry deposited N components into deposition increased the total N deposition at some sites in the Sierra Nevada and the Northeast by 100%, but there was great spatial variability. The contribution of dry materials to total N deposition to the Rockies was more muted and is consistent with a recent analysis of reactive N to Rocky Mountain National Park that attributed 15% of total N to dry deposition (mostly as NH_3^+; Beem et al. 2010).

The mean N deposition amounts for each of the three lake regions increased above those reported by NADP, as was expected, with the inclusion of topographically resolved precipitation (PRISM+NADP) and increased even more with the inclusion of dry deposition (total N) representation (table 2). The mean N deposition for the Sierra Nevada region increased from 1.5 kg N per ha calculated with NADP to 2.5 kg N per ha calculated with PRISM+NADP and further increased to 3.4 kg N per ha calculated with total N. For the Rocky Mountain region, the mean N deposition increased from 1.2 kg N per ha to 3.0 kg N per ha, then to 4.1 kg N per ha using the three calculation methods. For the Northeast, the mean N deposition from NADP was 4.7 kg N per ha, that from PRISM+NADP was 5.2 kg N per ha, and that from total N was 9.8 kg N per ha. Each of the three approaches toward estimating deposition has advantages and drawbacks, and because there is no one "right" method, we used all three for estimating critical loads for the lake regions.

Nitrogen deposition and lake nutrient enrichment. The "dog legs," or thresholds described above, where lake NO_3^- concentrations increased in response to N deposition, were determined by graphing the data. Although a more rigorous statistical approach would certainly be desirable, Dodds and colleagues (2010) noted considerable challenges and commented that the field of statistical threshold detection is in its infancy (Dodds et al. 2010). Both piecewise linear regressions and broken-stick models of lake NO_3^- concentrations by deposition using observed and natural-log-transformed data were conducted. The results were inconsistent across the regions. The statistical threshold identification matched the observations from the Rocky Mountains, but they were much higher than the observations for the other two regions and inconsistent with the definition of a critical load as the value below which there is no discernible change.

For the Rocky Mountains, the thresholds were crossed abruptly when deposition increased, reflecting the lack of watershed processes to attenuate nutrient inputs and the large number of lakes in the database (figure 5a–5c, table 2). The Sierra Nevada lakes also have low watershed vegetation, but with so few lakes, it was harder to discern a clear threshold. Nevertheless, concentrations increased with increasing deposition. The pattern for the Northeast reflected more of the "dog leg" described for European systems, perhaps because of the more complex forest and soil pathways tracked by incoming N deposition. Using NADP deposition, the thresholds (rounded to the nearest 0.5 kg N per ha per year) occurred at 1.5, 1.0, and 3.5 kg N per ha for the Sierra Nevada, the Rocky Mountains, and the Northeast, respectively. The threshold values for PRISM+NADP were 1.5, 2.0, and 3.5 kg N per ha, respectively, for these same regions. The deposition threshold for the Rocky Mountains doubled with the PRISM-enhanced deposition amounts. With total N deposition, the thresholds were 2.0 kg N per ha for the Sierra Nevada, 3.0 kg N per ha for the Rocky Mountains, and increased to 6.0 for the Northeast. Because both abiotic and biotic responses to N deposition in surface water begin only when there is excess NO_3^- above a reference value, the thresholds suggested in figure 5 and table 2 imply the lower limits, or critical loads, for N deposition, below which effects are unlikely (Stoddard et al. 2006).

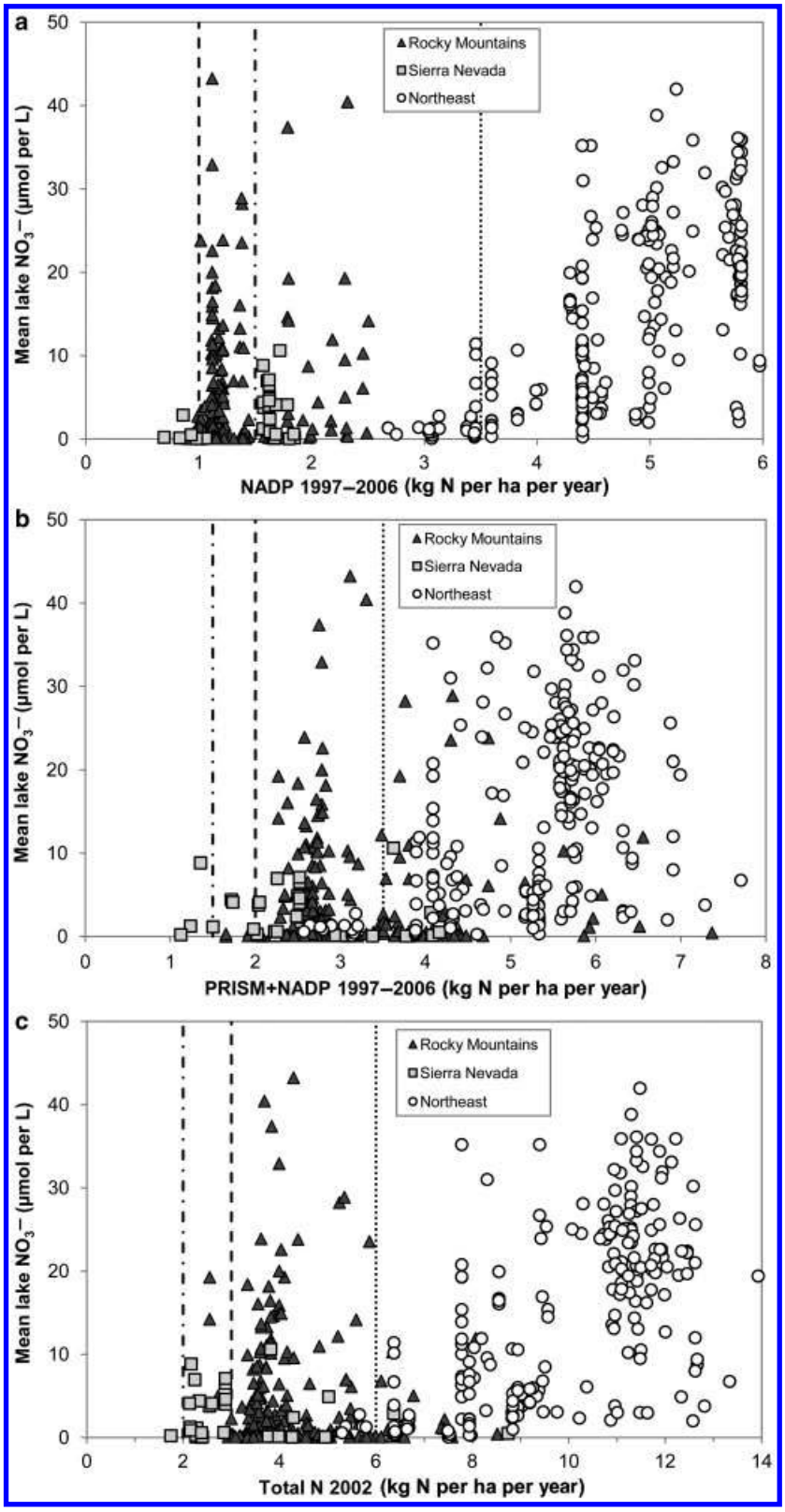

Figure 5. Mean NO_3^- concentrations of lakes versus mean nitrogen (N) deposition using (a) 1997–2006 National Atmospheric Deposition Program (NADP). (b) 1997–2006 mean PRISM (Parameter-elevation Regressions on Independent Slopes Model) precipitation and NADP wet N concentrations (PRISM+NADP). (c) 2002 total N. Thresholds were rounded to nearest 0.5 kilogram (kg) N per hectare (ha) per year and are dotted for Northeast, dashed for Rocky Mountains, and dashed-dotted for Sierra Nevada. Abbreviation: µmol per L, micromoles per liter.

The thresholds represent only conditions of N deposition above which elevated NO_3^- leaching has been noted at some sites and below which leaching rarely occurs. The relation between lake NO_3^- and N deposition does not hold at higher deposition amounts. The mechanism or mechanisms responsible for regional differences in the thresholds may be a function of catchment characteristics or land-disturbance history or an artifact of the few lakes in our data sets located in regions of high N deposition.

The N deposition at which lake NO_3^- concentrations were observed to increase suggests a preliminary nutrient enrichment critical load of 1.0 to 3.0 kg N per ha per year for the western mountains. Given the total-N-calculated range of N deposition in the United States of from less than 1.0 to 32.0 kg N per ha per year, these critical loads are low but comparable to the 1.5 kg N per ha per year proposed by Baron (2006) and to the 1.4 kg N ha per year proposed by Saros and colleagues (2010) and are only slightly lower than the 2.5 kg N per ha per year value from Bergström and Jansson (2006), who derived their results from biological response. The precautionary principle, suggested by Dodds and colleagues (2010) for the management of freshwaters, might warrant selection of critical loads of less than 2.0 kg N per ha per year, especially since the body of evidence from independently derived values from other studies led to similar conclusions.

The critical load for nutrient enrichment of lakes in forested catchments in the Northeast, derived from lake NO_3^- concentrations, ranged from 3.5 to 6.0 kg N per ha per year, depending on the N deposition method used. As was noted in table 1, there are some N-limited lakes in the East. For those lakes, elevated N atmospheric deposition may stimulate primary productivity and alter algal communities. However, although these ecosystems currently receive elevated N deposition, our ability to attribute biological changes directly to N deposition is confounded by decades of elevated atmospheric N deposition and land-use change.

Nitrogen deposition and lake acidification. The critical load of N for episodic acidification could be surpassed independently of the critical load for nutrient enrichment, as a result of physical and biological buffering mechanisms in catchments. There are few locations in the United States with sufficient sampling frequency to capture transient ANC depression caused by N deposition. We used those, and additionally estimated critical loads of N for episodic acidification using the literature and expert judgment.

There are no chronically acidic lakes in the West from acidic deposition, but Williams and Tonnessen (2000) observed N-induced spring acid episodes in a Colorado Front Range lake during snowmelt with annual wet N deposition rates of 4.0 kg N per ha per year. The value is within the range of the acidification thresholds of 4.0 to 8.0 kg N per ha per year for the Colorado Front Range, calculated using the Model of Acidification of Groundwater In Catchments (Sullivan et al. 2005).

Measured lake chemistry from long-term monitoring as part of the EPA Temporally Integrated Monitoring of Ecosystems (TIME) project (Stoddard et al. 1996) indicate that, as of 2008, about 6% of the lakes in the Northeast were chronically acidic (i.e., summer ANC < 0 μeq per L), and approximately 20% of the northeastern lakes had ANC values less than 10 μeq per L. We therefore estimated that a 10-μeq-per-L decrease in ANC during spring snowmelt or storm events would result in about 20% of the northeastern lakes experiencing acid episodes. We also estimated that a 10-μeq-per-L decrease in ANC corresponds to an episodic increase in NO_3^- of approximately 20 μeq per L, on the basis of the stoichiometric response of changes in ANC with additions in strong acid anions during snowmelt (Schaefer et al. 1990). TIME and other lake data assembled by Aber and colleagues (2003) showed that a lake NO_3^- of 20 μeq per L corresponded with 8 kg N per ha per year of atmospheric N deposition. We therefore suggest an N critical load for acidic episodes of approximately 8 kg N per ha per year, assuming that SO_4^{2-} concentrations remain constant (table 2; Aber et al. 2003).

Recovery from N-induced nutrient enrichment and acidification

Critical loads are used in the European Union and Canada as guides for air quality management. The loads are used to set goals for environmental protection. Although there is growing US evidence of recovery of some lakes from SO_2 emissions reductions in response to the Clean Air Act amendments, we have no such history yet for N deposition. The following section is therefore somewhat speculative but postulates what could be expected for protection against or recovery from acidification or nutrient enrichment N emissions reductions on the basis of the use of critical loads.

A scenario is easiest to describe for the western mountains, where the critical load of 4.0 kg per ha per year for acidification was exceeded in the data in only a few places (figure 5; Williams and Tonnesson 2000, Fenn et al. 2003). Atmospheric N deposition that exceeds the nutrient enrichment critical load of 1.0 to 3.0 kg N per ha per year is more widespread (figure 5). Restoration of low absolute N conditions in western lakes could therefore occur with N emission controls. Just as marked decreases in freshwater SO_4^{2-} concentrations and an increase in pH and ANC have occurred in the eastern United States and in Western Europe in response to SO_2 emissions reductions in the 1990s (Skjelkvåle et al. 2005), decreases in N emissions should result in decreases in NO_3^- and increases in ANC in western lakes. We speculate that oligotrophic phytoplankton assemblages should recover if nutrient sources are controlled but recommend experiments, similar to those conducted in Norway, to measure the biological and chemical reversibility of freshwater acidification (Wright et al. 1993).

It is more difficult to predict the effects on freshwaters of the northeastern United States from decreasing N emissions to the level of the critical load. In spite of decreases in SO_2 emissions, full recovery to preindustrial conditions has not occurred, in part because of continued atmospheric N deposition (Skjelkvåle et al. 2005). Decreases in N emissions should decrease lake NO_3^- and enhance the recovery of ANC beyond what has been observed to date, lowering both nutrient enrichment and episodic acidification. However, terrestrial N cycling and storage, depletion of base available cations, and in-stream and in-lake NO_3^- and NH_4^+ processing all influence lake NO_3^- and ANC, so responses may not be direct (Kelly et al. 1987, Bernhardt et al. 2005).

The trophic response of northeastern lakes to decreases in N deposition is unknown, but there is evidence of rearrangements in algal assemblages with increased ANC. Experimental liming of acid waters to mitigate acidity has brought about biological recovery of phytoplankton, periphyton, and zooplankton (Yan et al. 2003). Some acid-sensitive invertebrates and diatoms have naturally recolonized recovering freshwater lakes in Norway and Ontario (Raddum and Fjellheim 1984, Dixit et al. 2002). In Ontario lakes recovering from acidic deposition, Dixit and colleagues (2002) observed the return of acid-sensitive diatoms and chrysophytes as the pH of lakes increased; however, these were different taxa than those that had been present before the advent of acidic deposition. Factors that may limit the return of preindustrial aquatic communities even with reduced N deposition include the extirpation or extinction of taxa for recolonization and changing baseline conditions due to land disturbance, climate change, and atmospheric deposition (Dixit et al. 2002, Derry and Arnott 2007).

To the extent that N deposition contributes to soil and surface-water acidification, it can result in long-lasting changes to aquatic food webs. Precipitous calcium decline in the soft-water lakes of New England and southern Ontario, caused by acid deposition–induced soil base cation leaching, may contribute to the near extirpation of calcium-rich zooplankton (Jeziorski et al. 2008). Clearly, more research is needed on how and to what extent recovery of aquatic biota from acidification and N deposition will occur.

Conclusions

The establishment of a critical load should be treated as an exercise in adaptive management (and thus as a hypothesis to be reexamined). As more data and better statistical approaches become available, the values should be revised if necessary. Empirically derived critical loads are widely used in Europe but are augmented with steady-state and dynamic models (Hettelingh et al. 2009). Similar blending of empirical evidence with models for the United States is recommended. Used alone, the empirical approach to establishing critical loads for broad geographic areas has drawbacks, particularly for nutrient enrichment thresholds. High among these is the lack of a standard method for reporting deposition. There is also a shortage of ecosystem response data, whether experimental or from long-term monitoring, for most regions.

However, in spite of these concerns, we found great similarity in the nutrient enrichment critical load for the Sierra Nevada and the Rocky Mountains on the basis of the relationship between N deposition calculated three different ways and lake NO_3^- concentrations. Encouragingly, the critical load of 1.0 to 3.0 kg N per ha per year that we developed by relating deposition to lake chemistry (figure 5) matches those attained from biological hindcasting approaches and correlative studies. This very low N critical load is due to extreme topographic relief, shallow soils, sparse vegetation cover, and strong snowmelt control of hydrologic flushing in the watersheds surrounding western alpine lakes. Nitrogen deposition directly influences lake chemistry and biology with little to no lag time, and our confidence in empirically determined critical loads is fairly high. The mean lake NO_3^- concentrations, when compared with the range of NO_3^- known to stimulate productivity or alter algal assemblages, suggests that the N critical load of at least some lakes in the western mountains is exceeded.

The lowest empirically derived N deposition values in the Northeast were higher than those from the West. Independent biological measures for Northeast nutrient enrichment critical loads are lacking, and we do not know the extent to which our values of 3.5 to 6.0 kg N per ha per year are an artifact of current elevated N deposition, although that is certainly suggested from the modeled stream N concentrations of Smith and colleagues (2003). The deposition range also reflects the broader disparity between measured wet and total N deposition. The occurrence of N limitation in some northeastern lakes lends some credibility to our critical load estimates, but we recommend additional studies and modeling efforts.

Nitrogen-induced episodic acidification critical loads were higher than nutrient enrichment critical loads for western mountain and Northeast lakes. Here, a higher critical load value for the Northeast lakes than for the Sierra Nevada and the Rocky Mountain lakes seems appropriate. Forests and wetlands in the surrounding watersheds of Northeast lakes have greater N demand than western catchments with little upstream vegetation.

Nitrogen is one of a handful of major elements that sustain life on Earth. Elevated amounts of reactive, available N are now evident in many parts of the world. Although there can be benefits, including forest carbon sequestration, associated with elevated N inputs for some ecosystems, these conditions can result in undesirable nonlinear changes to natural ecosystems where native biodiversity is a priority. For the extremely responsive western alpine lakes in our sample set, even slight additions of N above background levels triggers nutrient enrichment with altered algal assemblages and increased productivity; our estimates of the thresholds, or critical loads, for nutrient enrichment are 1.0 to 3.0 kg N per ha per year and 3.5 to 6.0 kg N per ha per year for the western mountain and Northeast minimally disturbed lakes, respectively. Higher rates of N deposition (4.0 kg N per ha per year and 8.0 kg N per ha per year, respectively) lead to acid episodes. Air quality policies that reduce emissions in order to decrease deposition below these critical loads might lead to recovery of natural species assemblages in the Sierra Nevada and the Rocky Mountains. Recovery of ANC from many decades of acidic deposition in Northeast lakes will be improved by lower N deposition, and aquatic communities adapted to lower nutrient availability may replace current assemblages, but a return to preindustrial conditions may not be possible because of the loss of original taxa and continuing environmental change.

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