18 INLAND SURFACE WATERS

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18.1 Ecosystems Description

Freshwater aquatic ecosystems include rivers and streams, large and small lakes, reservoirs, and ephemeral ponds. Wetlands are defined and discussed in Chapter 17 of this report. It is estimated that there are 123,400 lakes with a surface area greater than 4 ha in the United States. Most lakes, however, are smaller than 4 ha; small lakes account for the majority of lake surface area both globally and in the United States (Table 18.1; Downing et al. 2006). The density of lakes varies greatly by region of the country, from 8.4 lakes per 100 km^2 in the upper Midwest and 7.8 lakes per 100 km^2 in Florida, to much lower values in other areas of the country (e.g., mid-Atlantic, Southeast, and West <1.0 lakes per 100 km^2) (Eilers and Selle 1991). The cumulative surface area of these lakes is approximately 9.5 million ha. The U.S. Geologic Survey's National Hydrographic Dataset (NHD) estimates that there are approximately 1.1 million km of perennial flowing streams in the United States. Of these about 91 percent are first through fourth order ("wadeable") (US EPA 2006).

18.2 Ecosystem Responses to N Deposition

A cascade of environmental effects occurs in response to elevated inputs of reactive N (Chapter 1; Galloway et al. 2003), but in freshwater ecosystems effects can be grouped under the categories of eutrophication (excess nutrient N) and acidification. Reactive N from anthropogenic activities enters lakes and streams from a number of sources, including wastewater treatment point sources, agricultural and other nonpoint sources, and atmospheric deposition (Driscoll et al. 2003b). The relative importance of these reactive N sources varies by the position of lakes and streams on the landscape. In protected headwater areas (such as national or state forests or parks), atmospheric deposition is the only source of anthropogenic reactive N. Surface waters with drainage from agricultural, industrial, or urban areas may receive proportionally less reactive N from atmospheric deposition and more from point and other

Table 18.1—Number and distribution of lakes (>4 ha in surface area) in the United States (after US EPA 2010)

| Lake size category *ha*	Total number of lakes in the U.S.	Cumulative Surface Area *ha*
1-4	155,201	310,402
4-10	68,559	433,605
10-20	24,902	352,167
20-50	16,488	521,396
50-100	6,134	433,739
>100	7,356	7,356,000
Total	278,640	9,407,310

nonpoint sources. The focus of this chapter is on surface waters that primarily receive N inputs from atmospheric deposition directly or indirectly through the watershed.

Freshwater aquatic ecosystems in which N has been observed to influence ecological processes either receive chronic or seasonally high inputs. Chemical responses range from long-term or temporary acidification (Dumont et al. 2005), to measurable N concentrations in the water column (Stoddard 1994). Nitrogen-limited waters respond rapidly to inputs of N with increased productivity, or eutrophication (Baron et al. 2000, Bergström and Jansson 2006).

The number of surface waters that are sensitive to or impacted by acidic deposition is relatively small compared to the total number of surface waters in the United States (Charles 1991). The areas of the United States with low values of acid neutralizing capacity (ANC <400 µeq/L) are shown in Figure 18.1. Of the areas shown, only those with ANC values <100 µeq/L would be considered sensitive to acidification by N deposition. These areas include the northern, central, and southern Appalachian Mountain regions, portions of southern New England, the Pine Barrens of New Jersey, portions of Georgia and Florida, portions of the upper Midwest, portions of the Rockies, the Cascades and the Sierra Nevada Mountains of the West. Note

Total Alkalinity of Surface Waters

by
James M. Omernik[1], Glenn E. Griffith[2],
Jeffrey T. Irish[2], and Colleen B. Johnson[2]
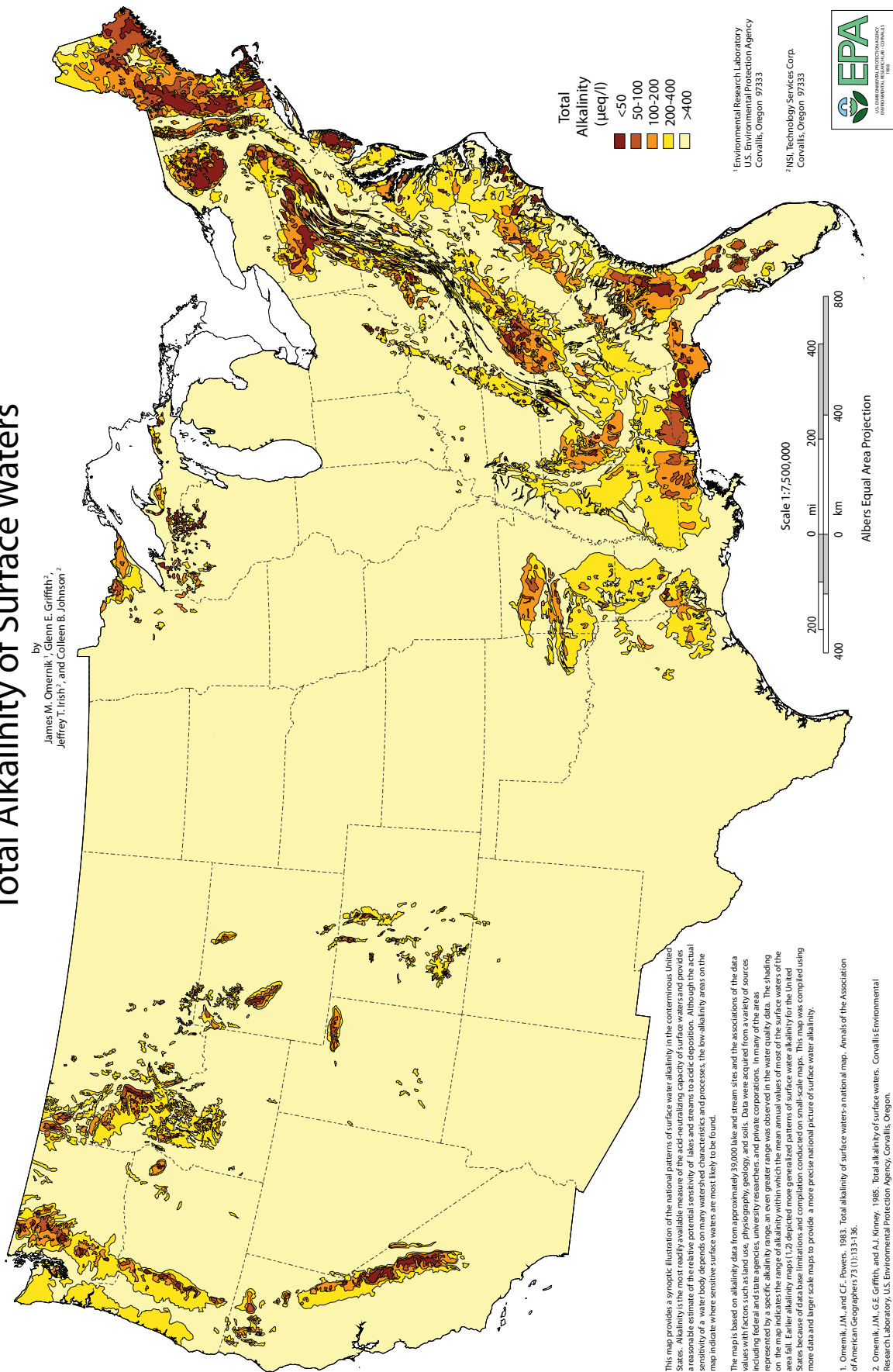
This map provides a synoptic illustration of the national patterns of surface water alkalinity in the conterminous United States. Alkalinity is the most readily available measure of the acid-neutralizing capacity of surface waters and provides a reasonable estimate of the relative potential sensitivity of lakes and streams to acidic deposition. Although the actual sensitivity of a water body depends on many watershed characteristics and processes, the low-alkalinity areas on the map indicate where sensitive surface waters are most likely to be found.

The map is based on alkalinity data from approximately 39,000 lake and stream sites and the associations of the data values with factors such as land use, physiography, geology, and soils. Data were acquired from a variety of sources including federal and state agencies, university researchers, and private corporations. In many of the areas represented by a specific alkalinity range, an even greater range was observed in the water quality data. The shading on the map indicates the range of alkalinity within which the mean annual values of most of the surface waters of the area fall. Earlier alkalinity maps (1,2) depicted more generalized patterns of surface water alkalinity for the United States because of data base limitations and compilation conducted on small-scale maps. This map was compiled using more data and larger scale maps to provide a more precise national picture of surface water alkalinity.

1. Omernik, J.M., and C.F. Powers. 1983. Total alkalinity of surface waters–a national map. Annals of the Association of American Geographers 73 (1):133-136.

2. Omernik, J.M., G.E. Griffith, and A.J. Kinney. 1985. Total alkalinity of surface waters. Corvallis Environmental Research Laboratory, U.S. Environmental Protection Agency, Corvallis, Oregon.

Total
Alkalinity
(µeq/l)
<50
50-100
100-200
200-400
>400

[1] Environmental Research Laboratory
U.S. Environmental Protection Agency
Corvallis, Oregon 97333

[2] NSI, Technology Services Corp.
Corvallis, Oregon 97333

Scale 1:7,500,000

Albers Equal Area Projection

EPA
U.S. ENVIRONMENTAL PROTECTION AGENCY
ENVIRONMENTAL RESEARCH LAB, CORVALLIS
1985

Figure 18.1—Map of surface water values of acid neutralizing capacity (ANC) across the U.S. Note that ANC values less than 100 µeq L^{-1} are more sensitive to the effects of acidic deposition (Omernik and Powers 1982).

that surface water acidification mostly results from the combined effects of atmospheric sulfur (S) and N deposition.

Episodic acidification from atmospheric N deposition has been reported mostly from snow-dominated environments where winter N deposition accumulates in the snowpack. Snowmelt triggers a pulse of reactive N to streams and lakes (Sickman and Melack 1998, Sickman et al. 2001, Stoddard 1994, Williams and Tonnessen 2000). Both preferential elution of solutes, including N, from the snowpack, and N-rich soil solutions produced by N mineralization and nitrification during the winter season of vegetation dormancy, are sources of spring nitrate (NO_3^-) pulses to streams and lakes (Brooks et al. 1998, Stoddard 1994). Episodic acidification associated with nitrate leaching can also occur during summer and fall rain events (Wigington et al. 1996).

The extent of lakes and streams affected by the eutrophication effects from atmospheric N deposition is much greater than those impacted by acidification (see below).

In the United States, atmospheric N deposition effects on surface waters have received less attention than effects from atmospheric S deposition. A number of factors have combined to draw the scientific and policy focus away from N. First, efforts to study the effects of atmospheric deposition had been largely directed toward surface water acidification, where the focus has been on chronic acidification, and particularly on the effects of S deposition on chronic acidification (Stoddard et al. 2003). The acidification response to N deposition is largely associated with episodic acidification, which has received less attention. Sulfur emissions and deposition have been declining since 1973, and with substantial decreases since 1995 following the 1990 amendments of the Clean Air Act (Dennis et al. 2007). In contrast, changes in atmospheric N deposition have been variable (Lehmann et al. 2005). Atmospheric NO_3^- deposition has generally decreased in the East following controls on emissions from electric utilities, but has increased in the west and central United States. Increases in ammonium (NH_4^+) deposition have been relatively widespread, particularly in the central and northern

Midwest (Lehmann et al. 2005, 2007). The relative importance of N deposition has increased in recent years as S deposition has decreased, creating an opportunity to re-examine the role of N deposition in surface water acidification (Lehmann et al. 2005).

Second, aquatic biologists have been focused on the role of phosphorus (P) in eutrophication of freshwaters for the past 40 years, largely due to the demonstrated effect of P increasing algal productivity worldwide (Schindler 1977, 1978). The prevailing wisdom is that P, an essential nutrient, is often the limiting nutrient to aquatic organisms, while N is tightly cycled in terrestrial systems, and that little N leaching occurs in undisturbed environments (Vitousek and Howarth 1991). Only within the past 20 years have there been studies questioning the established science and showing N limitation in some freshwaters, N leaching in some forest and alpine ecosystems, and N-induced acidification in lakes and streams with low ANC. A global meta-analysis of 990 freshwater field experiments found that N limitation of stream benthos, lake benthos, and phytoplankton was as common as P limitation from sites in all biomes worldwide (Elser et al. 2007); experimental additions of N to these ecosystems stimulated primary production. While P-amendment experiments in some lakes and mesocosms have shown an increase in compensatory N fixation, giving rise to the paradigm that P, not N, limits eutrophication (Schindler 1977, Schindler et al. 2008), many studies show N and P co-limitation (Elser et al. 1990, Elser et al. 2007). There presently is no evidence for a consistently significant contribution of planktonic N fixation to oligotrophic and mesotrophic lakes (Lewis and Wurtsbaugh 2008). Oligotrophic (nutrient poor) and ultra-oligotrophic waters are inherently sensitive to even low inputs of atmospheric N deposition (Bergström et al. 2005, Bergstrom and Jansson 2006, Elser et al. 2009, Lewis and Wurtsbaugh 2008).

The current extent of N limitation can be estimated from nutrient data collected as part of regional lake surveys in the eastern (Linthurst et al. 1986) and western (Landers et al. 1987) United States. Table 18.2 presents ratios of dissolved inorganic nitrogen (DIN; sum of NO_3^- and NH_4^+) to total phosphorus (TP)

Table 18.2—Proportion of lakes in three nutrient limitation classes (N limited, P limited, and N and P co-limited) based on ratios of dissolved inorganic nitrogen (DIN; NO_3^- + NH_4^+) to total phosphorus (TP). Data are from the Eastern Lake Survey (Linthurst et al. 1986), conducted in the fall of 1984, and the Western Lake Survey (Landers et al. 1987), conducted in the fall of 1985.

Region	Number of Lakes[a]	Number of N limited lakes (%)[b]	Number of P limited lakes (%)[c]	Number of lakes with co-limited N:P (%)[d]
Adirondacks	1,290	208 (16%)	777 (60%)	3.5 (23%)
New England	4,361	1,470 (34%)	1,363 (31%)	1,529 (35%)
Poconos/Catskills	1,506	607 (40%)	559 (37%)	341 (22%)
Southeast	2,424	735 (30%)	1,148 (47%)	541 (22%)
Upper Midwest	8,755	3,142 (37%)	2,787 (32%)	2,646 (31%)
Rockies	6,666	2,998 (45%)	2,259 (34%)	1,409 (21%)
Sierra/Cascades	4,155	1,468 (35%)	1,958 (47%)	729 (18%)
Total	28,976	10,627 (36%)	10,851 (37%)	7,499 (26%)

[a]The eastern and western Lake surveys were stratified random samples of lakes; 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 four were characterized as N limited, based on the work of Morris and Lewis (1988).

[c]Lakes with DIN:TP ratios (by weight) greater than 12 were characterized as P limited, based on the work of Morris and Lewis (1988).

[d]Lakes with DIN:TP ratios (by weight) between 4 and 12 could not be assigned to nutrient limitation class, and are characterized as either co-limited, or limited by something other than N or P.

as an indicator of nutrient status of lakes. These are based on water column analyses from the regional lake surveys. In Colorado lakes, Morris and Lewis (1988) found DIN:TP to be the most reliable predictor of nutrient limitation when compared against nutrient algal bioassays; they suggested using DIN:TP thresholds to identify probable N limitation (DIN:TP <4 by mass), P limitation (DIN:TP >12) and co-limitation (intermediate ratios). Percentages of N limited lakes vary from about 16 percent in the Adirondack Mountains, to 45 percent in the Rocky Mountains (Table 18.2). In general, N limitation is more common in regions with low N deposition (e.g., Sierra Nevada and Rocky Mountain ranges) and P limitation is more common in regions with higher N deposition (Adirondacks, New England, the Southeastern United States).

While the results in Table 18.2 suggest that N limitation is more widespread than is commonly understood, these values may actually underestimate historical rates of N limitation. Surveys of the literature report that oligotrophic waters are commonly N limited, especially undisturbed northern temperate or boreal

lakes that receive low levels of atmospheric N deposition (Bergström et al. 2005, Elser et al. 1990, Elser et al. 2007, Elser et al. 2009). A comprehensive study of available data from the northern hemisphere found unequivocal evidence of N limitation in lakes with low inputs of N, and increased N concentrations in lakes receiving N solely from atmospheric N deposition (Bergström and Jansson 2006). Studies of the response of phytoplankton to N or P amendments similarly found strong growth responses to N in lakes with low atmospheric N deposition, but no response to N in lakes with high N deposition (Elser et al. 2009). These authors suggested that the majority of lakes in the northern hemisphere may have originally been N limited, and that long-term atmospheric N deposition has changed the balance of N and P in lakes such that P limitation is observed today.

18.2.1 Abiotic Responses to N Deposition

The primary abiotic effect of excess N deposition for inland surface waters is acidification. Acidification occurs when acidic deposition (primarily sulfuric (H_2SO_4) and nitric (HNO_3) acids) exceeds the ability of watershed

soils, vegetation, and surface waters to neutralize these inputs. Catchment sensitivity to acidification is largely governed by bedrock and surficial geology, and much of the United States is insensitive to acidic deposition (Charles 1991). The regions of most concern for acidification are areas underlain by intrusive igneous rocks, typical of the western mountains, upper Midwest and parts of the Northeast, and base-poor sandstones typical of the Northeast, the mid-Atlantic, and the central and southern Appalachian Mountains (Figure 18.1; Charles 1991). Catchment sensitivity is also influenced by past acidic deposition, which can diminish the ability of soil to neutralize ongoing acidic deposition, due to long-term declines in exchangeable base cations (Driscoll et al. 2001, Huntington et al. 2000).

Surface water sensitivity to acidification is commonly measured in terms of pH and ANC. These indicators decline as lakes and streams acidify, and waters are classified as acidic when ANC values decrease to less than 0 µeq L^{-1}. Acidification can be further subdivided into chronic acidification (when lakes and streams are acidic year-round) and 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 can temporarily decrease ANC. While N deposition can influence both chronic and episodic acidity, it has the most marked effect on episodic acidification (Murdoch and Stoddard 1992, Schaefer et al. 1990, Wigington et al. 1996).

Severe episodes can produce conditions that are as deleterious to biota as chronic acidification (Baker et al. 1996). The mechanisms that produce acidic episodes include dilution, and flushing of NO$_3^-$, sulfate (SO$_4^{2-}$), and/or organic acids from forest soils or snowpack (Baker et al. 1996, Kahl et al. 1992, Lawrence 2002, Wigington et al. 1996, Williams and Tonnessen 2000). Acidic deposition may contribute to episodic acidification by supplying N to sensitive watersheds (producing pulses of NO$_3^-$ during high flow events), producing hydrologically mobile SO$_4^{2-}$, and by lowering baseline pH and ANC so that even modest episodes are sufficient to produce acidic (i.e., ANC <0 µeq L^{-1}) or near-acidic conditions (i.e., low but positive values of ANC). A generally accepted conceptual model of
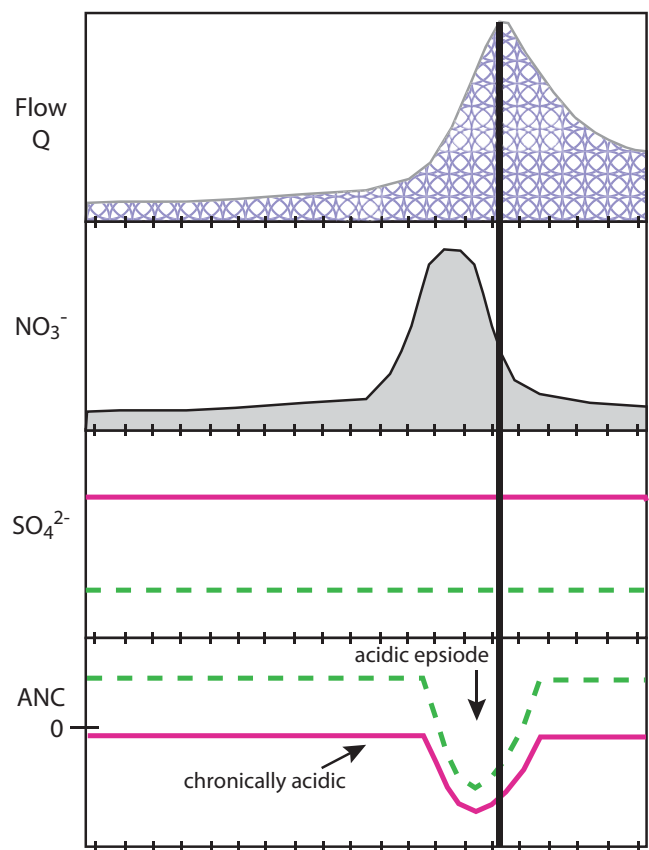

Figure 18.2—Conceptual diagram of changes in surface water chemistry of two contrasting upland streams impacted by atmospheric deposition in response to a hydrologic event. The upper two panels are discharge (Q) and nitrate (NO$_3^-$) during the stream event; the pattern is identical for both streams. The two streams have two different concentrations of SO$_4^{2-}$ in the third panel; the solid line represents a stream with high SO$_4^{2-}$ and the dashed line is a stream with low SO$_4^{2-}$. The fourth panel is acid neutralizing capacity (ANC). The stream with the higher SO$_4^{2-}$ (solid line) has a lower ANC during baseflow, and is chronically acidic. During the episodic event, ANC values decrease somewhat from low baseflow values, due to the pulsed input of NO$_3^-$. The low SO$_4^{2-}$ stream (dashed line) has low, but positive ANC during baseflow. At this site the pulse of NO$_3^-$ during the event causes a short-term decrease in ANC to acidic conditions (ANC < 0 µeq/L).

episodic acidification is that hydrologic events dilute base cation concentrations and trigger a pulsed increase in NO$_3^-$, causing short-term decreases in ANC (Figure 18.2). In low ANC waters, hydrologic events can result in decreases in ANC to near or below 0 µeq L^{-1}, causing adverse ecological effects (Baker et al. 1996). The deposition and associated leaching of SO$_4^{2-}$ lowers the ANC of surface waters during baseflow, so that short-term pulses of NO$_3^-$ (and, to a lesser extent, pulses of stored SO$_4^{2-}$ and naturally produced organic acids)

increase the severity of episodic or seasonal acidification (Driscoll et al. 2001, Stoddard et al. 2003, Wigington et al. 1996).

18.2.2 Biotic Responses to N Deposition

The biotic effects of excess N deposition for inland surface waters can be caused by both acidification and eutrophication. Acidification can have toxic effects on animals (zooplankton, macroinvertebrates, and fish), often as a result of the mobilization of aluminum and low pH conditions (Driscoll et al. 2001). Nutrient N primarily affects lower trophic levels (phytoplankton, periphyton) by increasing productivity in N limited systems (Wetzel 2001). Increased productivity can lead to secondary effects, including changes in the species composition or biodiversity of algae. Whether low or modest N enrichment influences animal populations is poorly understood.

Although chronically high acid levels stress aquatic life, acidic episodes are particularly harmful because abrupt, large changes in water chemistry allow fish and other organisms few areas of refuge (Baker et al. 1996). High concentrations of dissolved inorganic aluminum (Al) are directly toxic to fish, and pulses of Al during acidic episodes are a primary cause of fish mortality (Baker et al. 1996, MacAvoy and Bulger 1995, Van Sickle et al. 1996). Lethal effects are brought about by a combination of high acidity and Al concentrations, which disrupt the salt and water balance of blood in a fish (Driscoll et al. 2001), and respiratory stress caused by gill damage and mucous clogging of gills (Baker and Schofield 1982).

For the analysis of nutrient N effects, we relied on papers and studies that linked aquatic biological and ecological response to atmospheric deposition, but the results are consistent with laboratory or *in situ* dose-response studies and even land-use change studies. Essentially, the productivity of minimally disturbed aquatic ecosystems is often limited by the availability of N, and slight increases in available N trigger a rapid biological response that increases productivity and rearranges algal species assemblages (Lafrancois et al. 2004, Michel et al. 2006, Nydick et al. 2004, Saros et al. 2005). The mechanism for change is alteration of

N:P ratios, which can increase productivity of some species at the expense of others (Elser et al. 2009). Higher trophic levels (zooplankton, macroinvertebrates) may be secondarily affected by N, but further increases in primary, or autotrophic, production will be limited by other nutrients such as P or silica (Si).

18.3 Range of Responses Observed

18.3.1 Observed Abiotic Responses to N Deposition

Episodic acidification is more widespread than chronic acidification and can be important in acid-sensitive regions. For example, roughly 10 percent of Adirondack lakes were chronically acidified in the early 1990s, but more than three times that number (31 percent) underwent episodic acidification (Lawrence 2002). In a study of episodic acidification in the northeastern United States, Wigington et al. (1996) ranked the importance of various mechanisms in producing short-term acidity. Pulses of NO_3^- were ranked as the second most important process in the Adirondack and Catskill mountains; only dilution of base cations, which occurs in virtually all surface waters under increased flow, ranked higher.

In the western United States, there are no lakes that are chronically acidic, but episodic acidity has been observed in both the Colorado Front Range and Sierra Nevada Mountains, where episodic NO_3^- leaching during snowmelt decreases ANC to 0 µeq L^{-1} or below (Leydecker et al. 1999, Stoddard 1995, Williams and Tonnessen 2000).

18.3.2 Observed Biotic Responses to N Deposition

Algae are the aquatic biota most likely to show rapid eutrophication effects from N deposition. Eutrophication effects are most likely to occur in fresh waters that historically received low nutrient inputs. Especially in the western United States, these ecosystems are mainly located at high elevations. Studies have shown an increase in phytoplankton biomass in lakes with increasing N deposition in the Snowy Range in Wyoming, the Colorado Front Range, Sierra Nevada, Sweden, and across Europe (Table 18.3; Bergström et al. 2005, Bergström and Jansson 2006, Elser et al. 2009, Lafrancois et al. 2004, Nydick et al. 2004,

Table 18.3—Nitrate concentrations at which phytoplankton response has been observed

Site	N concentration $mg\ NO_3^--N\ L^{-1}$	Response	Study
Colorado Front Range	1.1	Diatom spp. growth stimulation	Lafrancois et al. 2004
Snowy Range, WY	1.0	Shift in species composition	Nydick et al. 2004
Beartooth Mountains WY	0.9	Stimulation of *A. formosa, F. crotonensis*	Saros et al. 2005
Sweden	0.9	Increasing biomass	Bergström et al. 2005
Europe and No. Amer. Lakes	0.9	Increasing biomass	Bergström and Jansson 2006

Table 18.4—Biological response observed in paleolimnological studies

Site	Year(s)	Response (method)	Study
Colorado Rocky Mountains	1950	Increased microbial activity, greater primary production, change in species composition (isotopic analysis)	Enders et al. 2008
	1950	Greater primary production, stimulation of *A. formosa, F. crotonensis*, shift in species assemblages (pigment analysis).	Das et al. 2005
	1950	Greater primary production, Stimulation of *A. formosa, F. crotonensis,* shift in species assemblages (taxonomy and cell counts)	Wolfe et al. 2001, 2003
	1950-1964	Threshold modeled at 1.5 kg N ha^{-1} yr^{-1}	Baron 2006
Beartooth Mountain Range, WY	1990	Greater primary production, stimulation of *A. formosa, F. crotonensis*, shift in species assemblages	Saros et al. 2003

Sickman et al. 2003). Paleolimnological reconstructions show a stronger eutrophication response in lakes where N deposition is higher than in lakes with lower N deposition (Das et al. 2005; Enders et al. 2008; Saros et al. 2003; Wolfe et al. 2001, 2003; Table 18.4).

Community shifts in phytoplankton were observed in the Snowy Range, with chrysophytes (golden algae) favored in lakes having lower N and cyanophytes (photosynthetic bacteria, also called blue-green algae) and chlorophytes (green algae) favored in lakes having higher N (Lafrancois et al. 2004). Sediment cores from lakes in the Colorado Front Range showed increasing representation of mesotrophic diatoms in recent times, as compared with predevelopment conditions (Enders et al. 2008; Wolfe et al. 2001, 2003). In Lake Tahoe, California, there has been a sharp increase in the ratio of araphidinate pennate (characteristic of nutrient rich conditions) to centric (characteristics of nutrient poor conditions) diatoms since about 1950 (largely due increases in *Fragilaria crotenensis),* associated with increased N loading to the lake. Jassby et al. (1994)

found that atmospheric deposition supplies most of the N to Lake Tahoe. While runoff from catchment streams supplies organic N to Lake Tahoe (Coats and Goldman 2001), a recent meta-analysis finds little relation between DON runoff and atmospheric N deposition (Pellerin et al. 2006).

Two species of diatom, *Asterionella formosa* and *Fragilaria crotonensis*, now dominate the flora of at least several alpine and montane Rocky Mountain lakes (Baron et al. 2000; Interlandi and Kilham 1998; Saros et al. 2003, 2005; Wolfe et al. 2001, 2003). The growth of *A. formosa* has been stimulated with N amendments during *in situ* incubations, using bioassays and mesocosms (6.4 to 1616.0 µmol L^{-1} N, McKnight et al. 1990; 76.0 µmol L^{-1}, Lafrancois et al. 2004; 18.0 µmol L^{-1}, Saros et al. 2005). *In situ* incubations in large lakes of Yellowstone National Park also stimulated *F. crotonensis* (Michel et al. 2006). The N requirements for *A. formosa* and *F. crotonensis* were determined to be 0.041 µmol L^{-1} and 0.006 µmol L^{-1}, respectively; higher concentrations stimulated growth (Michel et

al. 2006). Other species, including *Tetracyclus glans*, and *Staurosirella pinnata*, have shown negative growth responses to increased N (Michel et al. 2006). In studies of lake sediment diatom remains, typical oligotrophic species such as *Aulacoseria perglabra*, *Cyclotella steligera*, and *Achnanthes spp.* declined coincident with the rise in dominance of *A. formosa* and *F. crotonensis* (Wolfe et al. 2003)

Interlandi and Kilham (1998) demonstrated that maximum species diversity was maintained when N concentrations were low (<3.0 µmol L^{-1}) in lakes in the Yellowstone National Park (Wyoming, Montana) region. The implication is that species diversity declines with increasing availability of N. This finding complements the results of terrestrial studies that also showed a negative relationship between species diversity and N availability (Gilliam 2006, Suding et al. 2005).

Studies of Lake Erie found N to be seasonally, but only moderately, limiting to phytoplankton growth. Phytoplankton were far more responsive to additions of P, iron (Fe), or Si than N during different seasons (Moon and Carrick 2007, North et al. 2007). Studies of Lake Superior have documented a century-long increase in lake NO_3^- concentrations, which has produced a stoichiometric imbalance and strong P limitation (Finlay et al. 2007, Sterner et al. 2007).

There have been few studies on the effects of nutrient enrichment from N deposition on aquatic animals. Nydick et al. (2004) found that zooplankton density and biomass were altered by changes in nutrients in enclosure experiments, probably as a result of poor food quality. A whole-ecosystem experiment at the Bear Brook watershed, Maine simulated the effects of N and S deposition by means of experimental ammonium sulfate (($NH_4)_2SO_4$) addition over a 10-year period. Researchers found that elevated N inputs had minimal effect on stream detritus processing (Chadwick and Huryn 2005). They also found that N additions had no significant effect on stream macroinvertebrate secondary production or varying production by functional feeding groups. They concluded that climate-related variables, such as flow duration and litter inputs, controlled secondary production when N was not limiting.

Nitrogen deposition appears to have stimulated productivity and altered algal species assemblages at deposition rates of 1.5 to 2.5 kg N ha^{-1}yr^{-1} (Baron 2006, Bergström and Jansson 2006). A meta-analysis of lakes from 42 regions of Europe and North America concluded that atmospheric N deposition was responsible for increased concentrations of N in lake water, and elevated phytoplankton biomass (Bergström and Jansson 2006). Bergström and Jansson (2006) found a consistent pattern of nutrient limitation, showing N limitation for deposition below approximately 2.5 kg N ha^{-1}yr^{-1}, co-limitation of N and P for deposition between ~2.5 and 5.0 kg N ha^{-1}yr^{-1}, and P limitation in areas with N deposition greater than 5.0 kg N ha^{-1}yr^{-1}.

18.4 Critical Loads Estimates

18.4.1 Critical Loads for Abiotic Responses to N Deposition

Despite the difficulty of predicting current and future N behavior in lakes and streams, as discussed below (see section 18.6), both mechanistic (Aber and Driscoll 1997) and conceptual (Stoddard 1994) models of N saturation predict that elevated rates of N deposition will eventually produce elevated runoff concentrations of NO_3^- in relatively undisturbed catchments. Empirical cross-site watershed studies show the relationship between watershed N concentrations or loss as a function of atmospheric N deposition (Aber et al. 2003, Baron et al. 2000, Elser et al. 2009, Fenn et al. 2003, Sickman et al. 2002).

Such cross-site analyses were originally conducted for Europe (Dise and Wright 1995, 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 "threshold" of deposition. For the European data, the threshold is roughly 10 kg N ha^{-1}yr^{-1} (Wright et al. 2001).

We examined watershed NO_3^- concentrations as a function of total atmospheric N deposition for the eastern (Aber et al. 2003) and western United States (Sickman et al. 2002). Relationships are presented for annual average NO_3^- and peak springtime NO_3^-
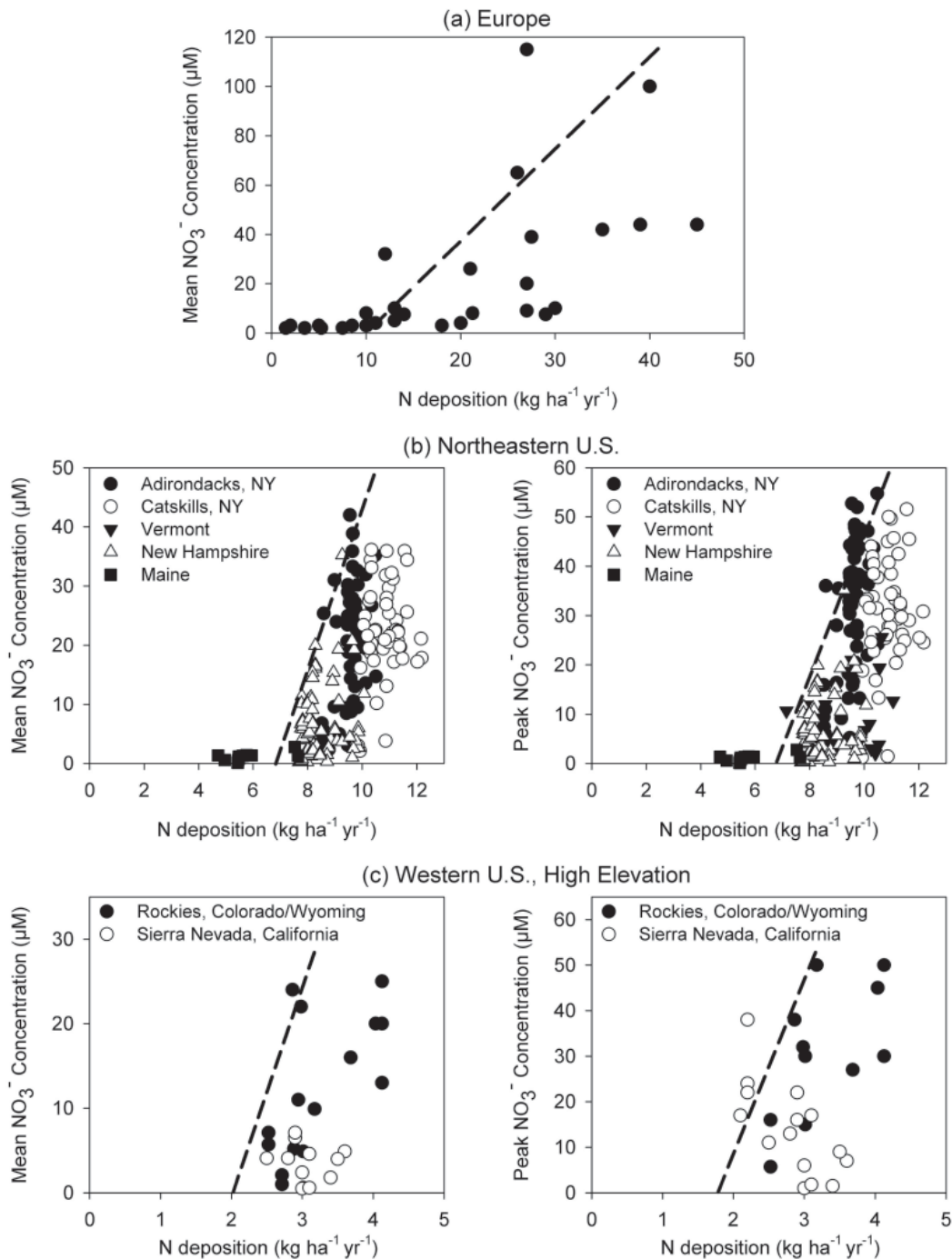
Figure 18.3—Relationship between mean and peak annual nitrate concentration (µM) and nitrogen deposition (kg N ha^{-1} yr^{-1}) in (a) forested catchments in Europe; (b) forested catchments in the northeastern United States; and (c) high elevation catchments in the western United States. European data from (Wright et al. 2001). Northeastern United States data are from Aber et al. (2003). Western NO$_3^-$ data are from Sickman et al. (2002) and Burns (2004). Western deposition was estimated from modeled NO$_3$ deposition (Nanus et al. 2003) and NH$_4^+$ deposition from the nearest NADP station. Lines are drawn by eye to estimate the x- intercept, the value of N deposition at which significant NO$_3^-$ leaching begins to occur.

in Figure 18.3. We consider the development of relationships between lake and stream NO$_3^-$ and atmospheric N deposition to be the first step toward empirical critical loads for N in these aquatic ecosystems. Because both abiotic and biotic responses to N deposition in surface water begin only when excess NO$_3^-$ begins to leach from catchments, the thresholds suggested in Figure 18.3 imply the lower limits for N

Table 18.5—Summary of critical loads of N for eutrophication and acidification for Eastern and Western high elevation lakes. Reliability rating: ## reliable; # fairly reliable; (#) expert judgement

	Western lakes $kg\ N\ ha^{-1}\ yr^{-1}$	Reliability for Western lakes	Eastern lakes $kg\ N\ ha^{-1}\ yr^{-1}$	Reliability for Eastern lakes
Critical load of N for eutrophication	2	##	8	#
Critical load of N for acidity	4-8	#	9	(##)

deposition below which effects are unlikely. The data suggest preliminary N critical loads of about 2 kg N ha^{-1}yr^{-1} for the western mountains, and 8 kg N ha^{-1}yr^{-1} for forested catchments in the East based on NO$_3^-$ leaching (Table 18.5). This value for the Western lakes can be considered a critical load of N for eutrophication. Note that there is some uncertainty in these estimates due to inherent difficulty in quantifying dry N deposition (see Chapter 3 this report). In addition, deposition varies by elevation within watersheds, adding additional uncertainty (Weathers et al. 2006).

We anticipate that the critical loads of N for acidity would be higher than the threshold values for eutrophication. Note that it is essential to establish the values of watershed ANC and SO$_4^{2-}$ and other major solutes in order to determine the critical load for acidity (Leydecker et al. 1999, Meixner et al. 2004). This is most effectively done with steady-state or dynamic acidification models that consider all major solutes simultaneously. We estimated critical loads of N for acidity using the literature and expert judgment. For Eastern lakes, we used TIME lake data (US EPA 2006) to identify lakes with low ANC values where a 10 µeq L^{-1} decrease in ANC, might increase the proportion of lakes exhibiting episodic acidification (ANC < 0 µeq L^{-1}). This change could result from an increase of approximately 20 µeq L^{-1} in NO$_3^-$ during snowmelt based on the stoichiometric response of changes in ANC with additions in strong acid anions during snowmelt (Schaefer et al. 1990). Comparing average NO$_3^-$ concentrations with snowmelt values for Eastern lakes (Figure 18.3), suggests an N critical load for acidity of approximately 9 kg N ha^{-1}yr^{-1}. This analysis assumes that SO$_4^{2-}$ concentrations remain constant and are not changing. This value is similar to that derived for Northern Forests (Chapter 7; Aber et al. 2003), as well

as that derived for European forested streams (Dise and Wright 1995), thus we consider it to be reliable.

Williams and Tonnessen (2000) reported episodic acidification in a Colorado Front Range lake during spring snowmelt with annual wet N deposition rates of 4 kg N ha^{-1}yr^{-1} (Table 18.6). An ecosystem modeling effort with MAGIC (Model of Acidification of Groundwater in Catchments) proposed an acidification critical load for the Colorado Front Range of 8 kg N ha^{-1} yr^{-1} (Table 18.6; Sullivan et al. 2005). Because the empirical value came from a high elevation lake that was highly responsive to changes in deposition values, while the modeled value was for a stream with at least some surrounding vegetation, we consider the measured value of 4 kg N ha^{-1}yr^{-1} to be fairly reliable, although more observations from additional lakes would lend strength to the critical load. Note these measurements are extremely difficult to obtain.

18.4.2 Critical Loads for Biotic Responses to N Deposition

Our estimate for N critical loads for eutrophication in the mountain West (i.e., 2 kg N ha^{-1} yr^{-1}) is similar to other estimates of aquatic critical loads for the region (Baron 2006, Fenn et al. 2003a). The estimated aquatic critical N load is lower than for terrestrial ecosystems, which is suggested to be 4 kg N ha^{-1} yr^{-1} for tundra based on experimental studies (Bowman et al. 2006), 3 to 4 kg N ha^{-1} yr^{-1} for tundra based on modeled studies (Baron et al. 1994), and 4 kg N ha^{-1} yr^{-1} for subalpine forest based on regional surveys (Rueth and Baron 2002). Nitrogen deposition appears to have stimulated productivity and altered algal species assemblages at deposition rates of 1.5 to 2.5 kg N ha^{-1} yr^{-1} (Table 18.7). A hindcasting exercise determined that the changes in algal communities in Rocky Mountain National Park

Table 18.6—Critical load for N for acidity for surface water in the Rocky Mountains. RMNP=Rocky Mountain National Park

Ecosystem	Site	Critical load for N deposition kg N ha^{-1} yr^{-1}	Response	Comments	Study
Alpine lake	Southern Rockies/Niwot	4.0	Episodic freshwater acidification	Observed	Williams and Tonnesson 2000
Alpine stream	Southern Rockies/Loch Vale RMNP	5	Episodic freshwater acidification	Modeled	Baron and Hartman 2004
Alpine stream	Southern Rockies/Loch Vale RMNP	7.3	Chronic freshwater acidification	Modeled	Baron and Hartman 2004
Alpine stream	Southern Rockies/Loch Vale RMNP	8	Freshwater acidification; ANC = 20 µeq L^{-1}	Modeled	Sullivan et al. 2005
Subalpine stream/lake	Southern Rockies/Loch Vale RMNP	12	Freshwater acidification; ANC = 20 µeq L^{-1}	Modeled	Sullivan et al. 2005
Alpine stream	Southern Rockies/Loch Vale RMNP	14	Freshwater acidification; ANC = 0 µeq L^{-1}	Modeled	Sullivan et al. 2005
Subalpine stream/lake	Southern Rockies/Loch Vale RMNP	21	Freshwater acidification; ANC = 0 µeq L^{-1}	Modeled	Sullivan et al. 2005

Table 18.7—Critical load for N for eutrophication for surface water in the Rocky Mountains. RMNP=Rocky Mountain National Park

Ecosystem	Site	Critical load for N deposition kg N ha^{-1} yr^{-1}	Response	Comments	Study
Alpine lake	Southern Rockies/ Loch Vale RMNP	1.5	Eutrophication	Paleolimnological	Baron 2006
Alpine lake	Northern Rockies/ Beartooth Mountains, WY	1.5	Eutrophication	Paleolimnological	Saros et al. 2003
Alpine stream	Southern Rockies/ Loch Vale RMNP	2	Freshwater eutrophication	Modeled	Baron et al. 1994
Alpine lakes	Rocky Mountains	2.5	Eutrophication, N and P co-limitation	Surveys and references therein	Bergström and Jansson 2006

that occurred between 1850 and 1964 were associated with an increase in wet N deposition that was 1.5 kg N ha^{-1} yr^{-1} (Baron 2006; Table 18.7). Similar changes inferred from lake sediment cores of the Beartooth Mountains of Wyoming also occurred at about 1.5 kg N ha^{-1} yr^{-1} deposition (Table 18.7). Pre-industrial inorganic N deposition is estimated to have been 0.1 to 0.7 kg N ha^{-1} yr^{-1}, based on measurements from remote parts of the world (Galloway et al. 2004, Holland et al. 1999). In the western United States, pre-industrial, or background, inorganic N deposition was estimated by Holland et al. (1999) to range from 0.4 to 0.7 kg N ha^{-1} yr^{-1}.

The determination of N critical loads for eutrophication of eastern lakes and the Great Lakes is problematic. Our analysis of N-limited lakes in lake districts of the United States suggests that, surprisingly, there are relatively large numbers of N-limited lakes in the East (Table 18.2). For those lakes, inputs of N associated with elevated atmospheric deposition are possibly stimulating plant productivity and altering algal communities, although mesotrophic lakes are often limited by P, and large lakes may be limited by Fe (Moon and Carrick 2007, Schindler et al. 2008, Sterner et al. 2007, Wetzel 2001). In a study of hundreds of Swedish lakes along

an N deposition gradient, lakes receiving greater than 4 kg N ha^{-1} yr^{-1} had greater ratios of chlorophyll a:total P, signifying a eutrophication response (Bergström et al. 2005). In the eastern United States, it is unknown whether a reduction in atmospheric N emissions and deposition would affect productivity in the N-limited lakes. Because these ecosystems are currently receiving elevated N deposition and have been receiving elevated deposition for decades, it is difficult to determine a value for critical loads of N. Moreover, evaluating the benefits associated with control on NO_3^- or NH_4^+ inputs in these N limited lakes is not straightforward. Inputs of NO_3^- and NH_4^+ are partially removed by in-lake processing (Kelly et al. 1987), which alters ANC. This process will influence the change in ANC associated with acidic deposition to eastern lakes.

18.5 Comparison to Critical Loads for Other Regions

The threshold of atmospheric N deposition at which elevated NO_3^- leaching may occur is highest in Europe, intermediate in eastern North America and lowest in the West (Figure 18.3). Note that elevated leaching losses do not necessarily occur at deposition levels above these thresholds. These thresholds only represent conditions of deposition above which elevated leaching has been noted at some sites and below which leaching rarely occurs. The mechanism(s) responsible for regional differences in the thresholds is not clear. Possible factors may include differences in the relative deposition of NH_4^+ and NO_3^-; more intensive forest and land management practices in Europe (Dise and Wright 1995), and differences in climate and hydrology. For a given region, even under elevated atmospheric N deposition, there is also considerable variability across sites. This variability may be due to several factors, the most important of which are likely land disturbance history (e.g., past forest cutting and agricultural practices) and climatic factors.

18.6 Future Research Directions and Limitations in Understanding

Adverse direct effects of N deposition on most fauna due to nutrient enrichment are probably minimal. Nitrogen concentrations alone are not high enough to influence animal metabolism, and the extent of eutrophication is insufficient (due to induced P limitation in oligotrophic waters) to cause oxygen depletion. Effects of N on invertebrates that are essential parts of aquatic food webs, however, are not well understood. Individual algal taxa are stimulated at low N concentrations, and several, including *Asterionella formosa* and *Fragilaria crotonensis*, have come to dominate some lakes that have received elevated N deposition (Saros et al. 2005, Wolfe et al. 2003). Other species have declined commensurate with increased available N, but we found only one study that addressed the question of effects of N deposition on freshwater aquatic diversity. Clearly, more work is needed to clarify these effects.

Research is needed to address two other questions. First, how do freshwater ecosystems respond to decreases in inputs of N? Alternative responses, which are not mutually exclusive, include decreases in productivity, reversion of species assemblages to former compositions, or irreversible alterations that develop into novel ecosystems (Hobbs et al. 2006, Schindler et al. 2008). Second, how does climate change interact with N deposition to influence aquatic productivity and assemblages?

One manifestation of limited understanding of ecosystem response to atmospheric N deposition is our inability to effectively explain the long-term temporal patterns of surface water N concentrations and loss in watershed ecosystems. Nitrate concentrations have been increasing in Lake Superior since at least the 1930s, but the cause remains unknown, and examination of stable N isotopes shows a more complex N-cycling story than can be explained by atmospheric N deposition alone (Finlay et al. 2007). In the mountain West, a few monitoring sites have shown increases in surface water NO_3^- attributable to increases in atmospheric N emissions and deposition (Jassby et al. 1994, Williams et al. 1996). Other sites have reported measurably high N concentrations since monitoring began in the 1980s (Baron et al. 2000, Fenn et al. 2003a). In the Northeast, however, investigators have reported declines in surface water concentrations and loss of NO_3^- in watersheds (Driscoll et al. 2003a, 2007; Goodale et al. 2003; Stoddard et al. 2003), with little change

in atmospheric N deposition. The mechanism(s) contributing to this decline in surface water NO_3^- in the Northeast is not clear but may be influenced by carbon dioxide-fertilization of forest vegetation, shifts in the composition of tree species, increases in in-stream N retention and/or changes in climate (Bernhardt et al. 2005, Driscoll et al. 2003b, Goodale et al. 2003, Thomas et al. 2010).

Ecosystem models, as mentioned above, are useful for augmenting empirical studies for deriving critical loads. This is especially true for addressing acidification critical loads, but ecosystem models have value for aiding in selection of critical loads for eutrophication where measurements are scarce. Ecosystem models, while widely used to represent N-cycling and effects, are still unable to represent some responses, reflecting an inadequate understanding of ecosystem processes and drivers. Although there are many ecosystem N-cycling models, there are flaws in all of them. For example, PnET-CN was developed to simulate N dynamics in forest ecosystems (Aber et al. 1997). While PnET-CN depicts many N-cycling processes, it over-predicts recent surface water NO_3^- in eastern forest watersheds and fails to capture the recent declines in NO_3^- concentrations (e.g., Aber et al. 2002). DayCent-Chem is another promising nutrient-cycling and geochemical model, but it also is not able to reproduce New England stream NO_3^- trends (Hartman et al. 2007, Hartman et al. 2009). Hydrochemical models such as the Alpine Hydrochemical Model (AHM) suffer from similar problems in reproducing chemical weathering or specific events (Meixner et al. 2004). As with many active ecosystem and hydrochemical models, however, AHM is constantly undergoing revisions and improvements (Molotch et al. 2008). Without effective simulation models, it will be difficult to develop critical loads for N and conduct comprehensive critical loads for acidity.

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