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HINDCASTING NITROGEN DEPOSITION TO DETERMINE AN ECOLOGICAL CRITICAL LOAD

JILL S. BARON[1]

U.S. Geological Survey, Natural Resource Ecology Laboratory, Colorado State University, Fort Collins, Colorado 80523-1499 USA

Abstract. Using an estimated background nitrogen (N) deposition value of 0.5 kg $N{\cdot}ha^{-1}{\cdot}yr^{-1}$ in 1900, and a 19-year record of measured values from Loch Vale (Colorado, USA; NADP site CO98), I reconstructed an N-deposition history using exponential equations that correlated well with EPA-reported NO_x emissions from Colorado and from the sum of emissions of 11 western states. The mean wet N-deposition values for the period 1950–1964 was ~1.5 kg $N{\cdot}ha^{-1}{\cdot}yr^{-1}$, corresponding to the reported time of alteration of diatom assemblages attributed to N deposition in alpine lakes in Rocky Mountain National Park (USA). This value becomes the critical load defining the threshold for ecological change from eutrophication. Thus if an N-deposition threshold for ecological change can be identified, and the date at which that threshold was crossed is known, hindcasting can derive the amount of atmospheric deposition at the time of change, at least for alpine lakes. Independent support for the technique and the deposition amount comes from experimental studies, ecosystem modeling, and paleolimnological records from northern Wyoming (USA).

Key words: alpine lakes; atmospheric nitrogen deposition; critical load; ecological-change threshold; hindcasting; Loch Vale, Colorado, USA; Rocky Mountain National Park, USA.

INTRODUCTION

Wet and dry deposition of nitrogen (N) species have been measured for 25 years or less in the United States, making it impossible to know with certainty atmospheric deposition amounts from earlier in the 20th century. The post-1950 increase in emissions of reactive nitrogen to the atmosphere, specifically ammonia and nitrogen oxides, is a major disruption of the global nitrogen cycle (Galloway et al. 1995). Ecological ramifications, including increased forest and grassland productivity, eutrophication and acidification of freshwaters, hypoxia, and loss of biodiversity, have been documented in terrestrial, freshwater, and coastal ecosystems worldwide (Vitousek et al. 1997). Premeasurement deposition values have interest for scientists attempting to understand nutrient-cycling processes and ecological response, and also for policy makers who strive to protect ecosystems from undesirable environmental change. An estimate of the amount of N deposition at the time of environmental change could be used to define an ecological threshold, or critical load, beyond which ecosystem properties or characteristics differ from before (Porter et al. 2005). Such empirical knowledge of cause and effect could be used to predict future condition in regions where N deposition is still low, or used as a target for ecological restoration.

In the western United States pre-industrial, or background, inorganic-N deposition has been estimated at between 0.4 and 0.7 $kg{\cdot}ha^{-1}{\cdot}yr^{-1}$ (Holland et al. 1999). Current western inorganic-N deposition values are spatially heterogeneous, and range widely from 1.0–4.0 kg N (NO_3-N plus NH_4-N)$\cdot ha^{-1}\cdot yr^{-1}$ over much of the region to as high as 30.0–90.0 kg $N{\cdot}ha^{-1}{\cdot}yr^{-1}$ in heavily populated southern California (Fenn et al. 2003*b*). High inorganic-N deposition occurs at sites proximal to large sources of N emissions, such as highly urbanized or agricultural regions, and at high elevations, where orographic conditions contribute increased amounts of precipitation (Marner and Harrison 2004, NADP 2004). Current values in the western United States are at least an order of magnitude greater than background deposition values.

In the Colorado Front Range mountains observations of increased foliar and soil N, alpine plant-community changes, heightened rates of microbial mineralization and nitrification, elevated surface-water NO_3 concentrations, and changes in lake algal species assemblages have all been attributed to atmospheric N deposition (Baron et al. 2000, Korb and Ranker 2001, Nydick et al.

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[1] E-mail: jill_baron@usgs.gov

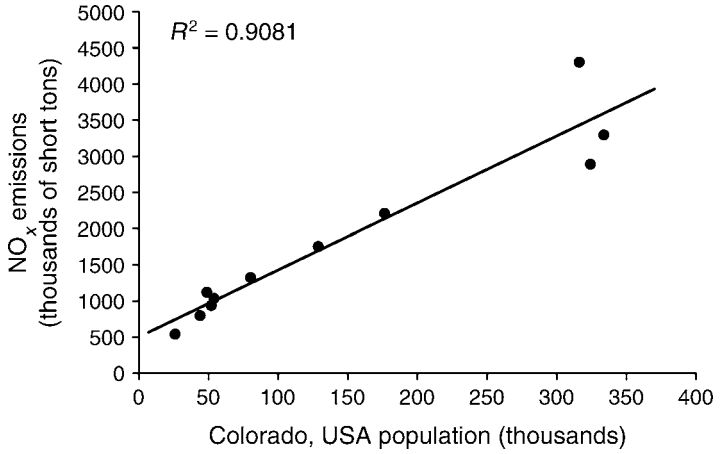

FIG. 1. Regression of Colorado population with Colorado NO_x emissions, in thousands of short tons (SI conversion: 1 short ton = 907.1847 kg).

2004). Further, sediment records from four alpine lakes in Rocky Mountain National Park within a 30-km radius reflect abrupt changes in algal diatom communities between 1950 and 1964 from typical undisturbed oligotrophic alpine lake flora to taxa associated with increases in inorganic-N availability (Wolfe et al. 2003).

The critical-load concept is used in many countries to integrate the effects of air pollution on ecosystems by quantifying an ecological threshold (Porter et al. 2005). A "critical load" can be defined as the amount of one or more pollutants that an ecosystem can safely absorb before there is a change in ecosystem state and/or in a particular ecosystem function. Williams and Tonnessen (2000) selected a critical load related to surface-water acidification for the Colorado Front Range of 4.0 kg $N \cdot ha^{-1} \cdot yr^{-1}$ based on alpine lake chemical responses to snowmelt. Nutrient enrichment, however, or eutrophication, is a fundamentally different type of environmental response, and occurs before acidification and at lower N-deposition amounts in nutrient poor environments (Fenn et al. 2003*a*). Protection from eutrophication requires establishment of a lower critical load. Since algae respond quickly to changes in their chemical environment (Charles and Whitehead 1986), the amount of atmospheric N deposition in the 1950s and 1960s that occurred in the Colorado Front Range might reflect the threshold, or critical load, above which ecological change occurred.

In this paper I present a simple method for hindcasting deposition trends in order to derive the critical load. On the premise that what goes up must come down, deposition patterns should mimic emissions patterns, allowing a back-calculation based on the relationship between post-1980 emissions and deposition (Howarth et al. 2002, Butler et al. 2003). However, the link between N emissions and deposition is not direct. Local climate variability, variation over time in the heights and mixture of reactive N compounds emitted or transported, and changes in vegetation type or landscape fragmentation can alter site-specific rates and amounts of deposition (Weathers et al. 2000, Butler et al. 2003, Marner and Harrison 2004). Further, there are sources of error and uncertainty in the emissions-inventory process (Majeed 2001). Methods for estimating reactive N emissions issued to the states by the U.S. Environmental Protection Agency (EPA) are not only complex, and thus subject to error, they have changed over time, with the most recent change in methods in 1997 (EPA 1997*a*, *b*). When comparing emissions to deposition, therefore, there may be sizeable uncertainty in values from both ends. Nevertheless, both emissions and deposition have increased over time (Galloway and Cowling 2002).

Nitrogen oxide (NO_x) emissions increased exponentially in Colorado and the western United States from 1900 to the late 1980s, and have been approximately stable since then (EPA National Emissions Inventory 2004).[2] The emissions increase has been concurrent with rapid population growth, and the linear correspondence of population increases with both western NO_x emissions and Colorado NO_x emissions is quite strong (Fig. 1; $R^2 = 0.91$). Similar relations between population and emissions worldwide have been noted by others (Galloway et al. 1995, Vitousek et al. 1997).

Ammonia emissions trends have not been estimated back to 1900 by EPA, and the accuracy of the NH_3 inventory is highly uncertain (Fenn et al. 2003*b*). Ammonia comes primarily from crop agriculture and livestock; irrigated agriculture and industrial animal-feeding operations have increased sharply along the Colorado Front Range since 1950 (Baron et al. 2000). Ammonium in wet atmospheric deposition has increased at 65% of western National Atmospheric Deposition Program sites since 1985 (Clow et al. 2003).

METHODS

The Loch Vale National Atmospheric Deposition Program (NADP) site, CO98, is located at 3159 m in a subalpine forest clearing in Rocky Mountain National

[2] ⟨http://www.epa.gov/air/data/⟩

Park (USA). Annual inorganic wet nitrogen concentrations (in milligrams of N per liter) of NO_3 and NH_4 were obtained from NADP.[3] Loch Vale data from calendar year 1988 were excluded due to four months of missing data. Annual inorganic wet N deposition was calculated by multiplying measured NO_3-N and NH_4-N concentrations with measured precipitation from NADP. Premeasurement NO_3-N and NH_4-N concentrations were extrapolated from measured concentrations using both linear and exponential functions with least-squares regressions (Microsoft Excel 2002). Premeasurement NO_3-N and NH_4-N depositions were calculated by multiplying both the linearly and exponentially estimated concentrations with two methods of estimating past precipitation: the mean annual 1984–2002 annual precipitation (1055 mm); and VEMAP-derived precipitation for 1900–1983, described below (Kittel et al. 1996). A preindustrial "anchor" deposition value of 0.5 kg $N{\cdot}ha^{-1}{\cdot}yr^{-1}$ was set for 1900 (Holland et al. 1999). Of that, 80% was assigned as NO_3-N, and 20% was assigned as NH_4-N, after Holland et al. (1999). VEMAP (Vegetation/Ecosystem Modeling and Analysis Project) data include a high-resolution topographically adjusted climate history of the United States from 1895 to 1993 on a 0.5° grid. The mean (and SD) for VEMAP precipitation for the Estes Park grid cell (covering Loch Vale) was computed for years 1900–1993. For each year,

$$\text{VEMAP}_i - \text{VEMAP}_\text{M}/\text{VEMAP}_\text{SD} = \text{standard deviation fraction}_i \quad (1)$$

where VEMAP_i is the annual precipitation for year i, $i =$ 1900–1993, and VEMAP_M and VEMAP_SD are the mean and standard deviation VEMAP precipitation, respectively. The precipitation sequence was adjusted for Loch Vale, for each year:

$$\text{SITE}_i = \text{SITE}_\text{M} + (\text{SITE}_\text{SD})(\text{standard deviation fraction}_i) \quad (2)$$

where SITE_i is the annual precipitation for Loch Vale, i = 1900–1993. Each VEMAP standard-deviation fraction was multiplied by the Loch Vale NADP standard deviation (SITE_SD), and added to the site's mean precipitation (SITE_M). All calculations were extrapolated backward from the beginning of 1984, the first full year of recorded measurements.

Dry-deposition flux data (1995–2002) were obtained from Clean Air Status and Trends Network (CASTNET)[4] site ROM406, located at 2743 m in Rocky Mountain National Park, 10 km from the Loch Vale NADP site.

Deposition time series were compared with EPA emissions estimates for 11 western states (Washington, Oregon, California, Idaho, Nevada, Utah, Arizona, Montana, Wyoming, Colorado, and New Mexico). National and individual state 1900–1970 NO_x emissions trends were estimated by EPA (EPA 1998).[5] Post-1970 estimates were from EPA National Emissions Inventories (EPA NEI 2004; see footnote 2). With 1991–1995 EPA emissions data and 1988 NADP data missing, there were only 12 pairs of emissions and deposition values.

TABLE 1. Loch Vale (Rocky Mountain National Park, Colorado, USA) precipitation characteristics, measured at the National Atmospheric Deposition Program site CO98.

Characteristic	Loch Vale (CO98)
Years of measured wet deposition	1984–2003
Site elevation	3159 m
Annual precipitation†	1054.8 ± 180.9 mm
Wet NO_3-N†	0.17 ± 0.02 mg/L
Wet NH_4-N†	0.11 ± 0.03 mg/L
Mean NH_4:NO_3	0.65
NH_4:NO_3 trend line	$y = 0.011x + 0.494$
Wet N deposition (kg/ha)†	2.94 ± 0.45 kg/ha
Annual dry N deposition†,‡	0.94 ± 0.26 kg/ha

† Data are means ± SD.
‡ Data from CASTNET (Clean Air Status and Trends Network) site ROM 406 ($n = 8$ years).

RESULTS

Examination of measured N

Mean annual wet NO_3-N and NH_4-N concentrations were 0.17 mg/L, and 0.11 mg/L^{-1}, respectively, at Loch Vale (Colorado, USA) for the period 1984–2002 (Table 1). The NH_4:NO_3 ratio increased from 1984 at a rate of 0.01 yr^{-1}, with a range of 0.46–0.88, and mean of 0.65. The NADP mean wet inorganic deposition was 2.94 ± 0.45 kg $N{\cdot}ha^{-1}{\cdot}yr^{-1}$ (Table 1, Fig. 2A). Dry N deposition (sum of NH_3-N, NO_3-N, and NH_4-N) was 20–50% of total wet N deposition, but annual wet and dry deposition values from the ROM406 CASTNET site correlated poorly ($R^2 < 0.1$; Fig. 2A). Because of dissimilarity in wet and dry inorganic-N deposition trends, and the different locations of dry and wet sampling sites, dry deposition values were not considered further.

Wet precipitation inorganic-nitrogen (NH_4-N plus NO_3-N) concentrations increased by 0.01 mg $N{\cdot}L^{-1}{\cdot}yr^{-1}$ between 1984 and 2002 (Fig. 2B). Year-to-year variability caused both linear ($R^2 = 0.36$) and exponential ($R^2 = 0.34$) equations to have poor goodness of fit. Whereas N concentrations increased steadily over time, the pattern for wet N deposition was more complex. High wet N-deposition years generally corresponded with years of higher precipitation, but not always. Wet N deposition during 1999–2001 was greater than 3 kg $N{\cdot}ha^{-1}{\cdot}yr^{-1}$ even though precipitation was less than 1000 mm/yr (Fig. 2A).

[3] ⟨http://nadp.sws.uiuc.edu/⟩

[4] ⟨http://www.epa.gov/castnet/⟩

[5] ⟨http://www.epa.gov/ttn/chief/trends/procedures/trends_procedures_old.pdf⟩

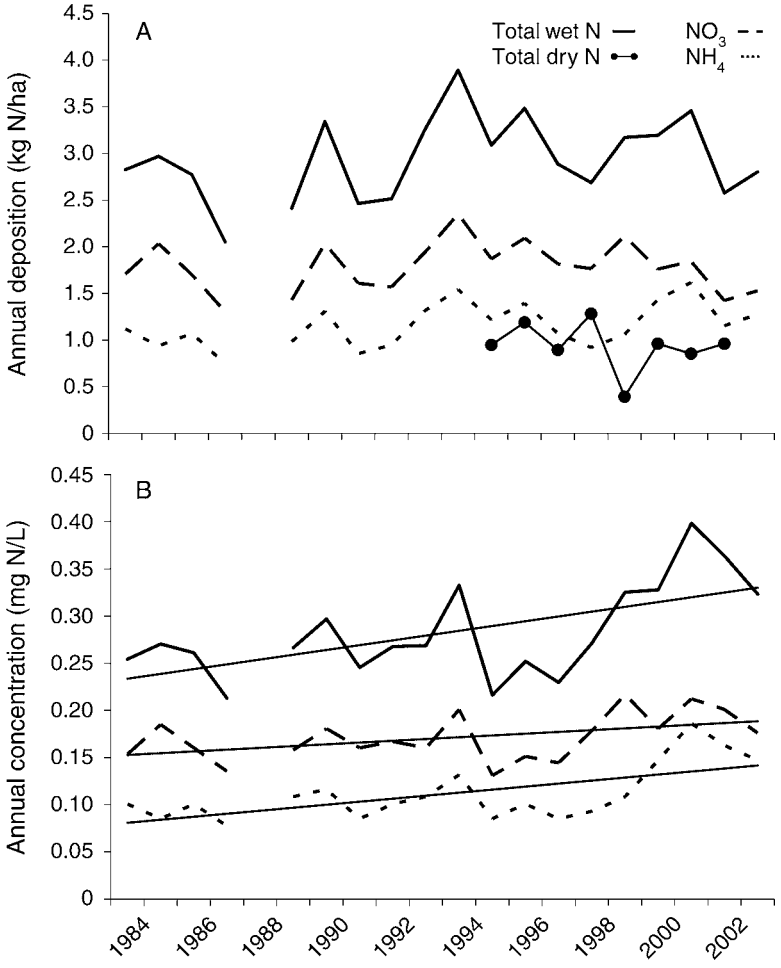

FIG. 2. N deposition data and concentrations (Colorado, USA), 1984–2002. (A) Annual deposition of wet inorganic N, wet NO_3-N, wet NH_4-N, and dry N (sum of NH_3-N, NO_3-N, and NH_4-N). Deposition data are from NADP site Loch Vale (CO98) and CASTNET site ROM406 in Rocky Mountain National Park. (B) Annual volume-weighted mean concentrations of wet inorganic N, wet NO_3-N, and wet NH_4-N (from Loch Vale). Data from 1988 were incomplete. The slopes of the trendlines are 0.005, 0.002, and 0.003 for N, NO_3-N, and NH_4-N, respectively.

Hindcasting deposition

All equations, by having a fixed endpoint in the year 1900, gave similar deposition hindcast curves (Fig. 3). Linear hindcasts of measured deposition gave the highest estimates of wet N-deposition values mid-century relative to the other methods, deviating from VEMAP-generated wet N-deposition values by up to 1.0 kg $N{\cdot}ha^{-1}{\cdot}yr^{-1}$ (Fig. 3A). Exponential hindcasts using mean precipitation were a smoothed version of VEMAP-generated deposition. Exponential hindcasts of measured deposition were identical to those calculated with mean precipitation. VEMAP-generated wet N deposition showed less interannual variability ($<$1.0 kg $N{\cdot}ha^{-1}{\cdot}yr^{-1}$) than post-1984 measured deposition variability, which ranged up to 1.4 kg $N{\cdot}ha^{-1}{\cdot}yr^{-1}$. The VEMAP-generated hindcast gives a conservative estimate of the range of deposition that could have occurred due to variability in precipitation.

The linear, exponential, and VEMAP-generated curves for wet NO_3-N deposition were similar to those for total wet N (Fig. 3B). Ammonium-deposition hindcasts depicted very low amounts early in the century, increasing to make up nearly 40% of total inorganic wet N deposition by 1982 (Fig. 3C).

Relation between deposition and NO_x emissions

There was no significant relationship between measured concentrations, measured deposition, or emissions at Loch Vale; R^2 values ranged from 0.05 to 0.14. Correlations among these variables improved markedly when 100 years of emissions estimates were compared with back-calculated wet depositions (Table 2). The R^2 values for total inorganic wet N deposition and wet NO_3-N deposition with emissions had a range of 0.77–0.91, with higher correlations of wet deposition with emissions from all western states than from CO emissions alone.

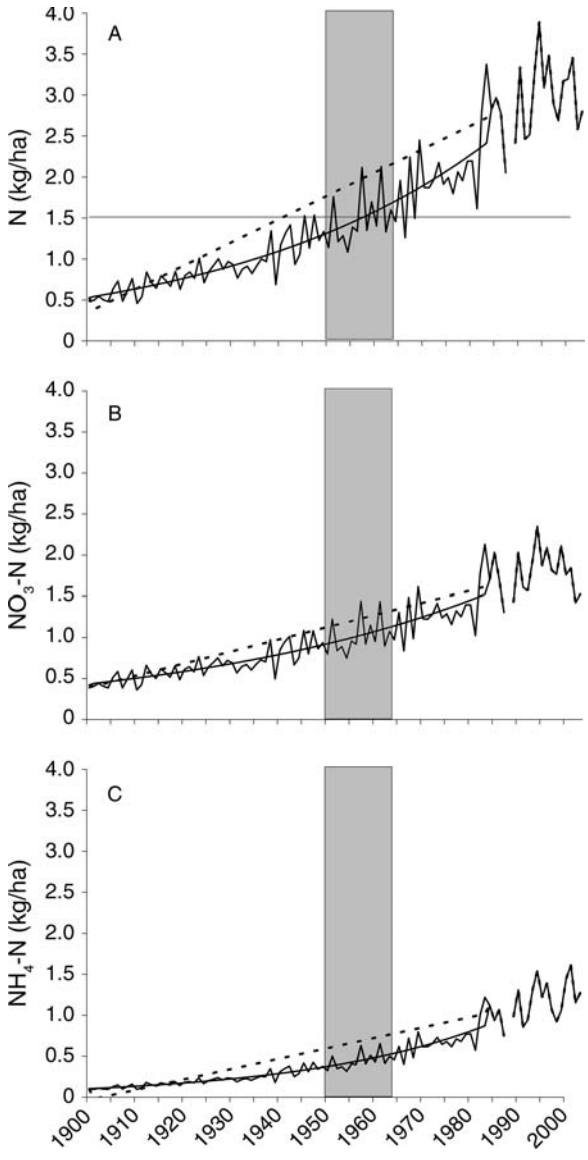

FIG. 3. Wet N-deposition trends, hindcast to 1900, of (A) wet N, (B) wet NO_3-N, and (C) wet NH_4-N. The broken lines indicate the linear extrapolation of annual volume-weighted mean (VWM) concentrations and 1984–2003 mean precipitation; smooth solid lines indicate the exponential equation of annual VWM concentrations and 1984–2003 mean precipitation; and variable solid lines indicate the exponential equation of VEMAP-generated precipitation and VWM concentrations. Actual 1984–2003 depositions are shown. The 1950–1964 period during which lake trophic state changed is shaded gray. In (A) the horizontal line at 1.5 kg N/ha is the mean 1950–1964 N deposition calculated from exponential VWM and VEMAP-generated hindcasts.

NO_x emissions were not correlated with wet NH_4-N deposition values. The R^2 values for total inorganic wet N, NO_3-N, and NH_4-N deposition with Colorado population ranged from 0.85 to 0.97. The correlations of hindcasts with population trends from all 11 western states were even greater (0.91–0.98).

DISCUSSION

Measured wet N concentrations increased since the 1980s, and the exponential trajectories created by hindcasting were very similar to historical emissions trends and population growth numbers. The exponential hindcast equations can logically be used to estimate a range of possible deposition values. Paleolimnological proxies of the onset of ecological change indicate diatom assemblages switched from ultra-oligotrophic to mesotrophic between 1950 and 1964 in four Rocky Mountain National Park lakes located within 30 km of the Loch Vale (Colorado, USA) NADP (National Acid Deposition Program) site (Wolfe et al. 2003). Wet N depositions between 1950 and 1964 were 1.51 ± 0.12 and 1.49 ± 0.32 kg $N \cdot ha^{-1} \cdot yr^{-1}$ (mean ± SD) for exponential and VEMAP equations, respectively (Table 3, Fig. 3A**)**. The critical load for eutrophication, therefore, falls around 1.5 kg wet $N \cdot ha^{-1} \cdot yr^{-1}$.

The link between deposition and aquatic ecological effects, which is important to establishing a critical load, must be site specific, since watershed characteristics influence how much of the wet N that is deposited makes its way to the lake. In the Rocky Mountains, lake N concentrations are inversely related to the amount of upstream soil cover (Sickman et al. 2003). For alpine headwater lakes where there is little surrounding vegetation the hindcasting method may provide a reasonable method for linking deposition to environmental change. There are several lines of evidence suggesting a critical load of 1.5 kg wet $N \cdot ha^{-1} \cdot yr^{-1}$ may be appropriate.

Mesocosm studies that raised N concentrations to 1.0 mg N/L brought about shifts in phytoplankton composition in nutrient-poor Wyoming (USA) lakes (Nydick et al. 2004), similar to other studies (Cottingham et al.

TABLE 2. Correlations (R^2) between hindcasts of wet inorganic-N, wet NO_3-N, and wet NH_4-N deposition at Loch Vale (Colorado, USA) with estimated NO_x emissions and population from 1900 to 2000, for Colorado and for 11 total western states.

Regression models	NO_x emissions		Population	
	Colorado	Western states	Colorado	Western states
N				
Linear	0.80	0.90	0.89	0.95
Exponential	0.89	0.90	0.94	0.97
VEMAP	0.80	0.81	0.94	0.95
NO_3-N				
Linear	0.80	0.90	0.85	0.91
Exponential	0.88	0.91	0.90	0.94
VEMAP	0.77	0.80	0.89	0.92
NH_4-N				
Linear	n.a.	n.a.	0.94	0.98
Exponential	n.a.	n.a.	0.97	0.97
VEMAP	n.a.	n.a.	0.96	0.95

Note: VEMAP = Vegetation/Ecosystem Modeling and Analysis Project; n.a. = not applicable.

TABLE 3. Wet atmospheric inorganic-N, NO_3-N, and NH_4-N concentrations and deposition at Loch Vale (Colorado, USA) from hindcasts, 1950–1964.

N source	Range	Mean (SD)
Inorganic N		
Concentration (mg/L)		
Linear	0.17–0.21	0.19 (0.01)
Exponential	0.13–0.16	0.14 (0.01)
Deposition (kg/ha)		
Linear	1.78–2.18	1.98 (0.13)
Exponential	1.32–1.71	1.51 (0.12)
VEMAP	1.08–2.13	1.49 (0.32)
NO_3-N		
Concentration (mg/L)		
Linear	0.11–0.13	0.12 (0.01)
Exponential	0.09–0.11	0.10 (0.01)
Deposition (kg/ha)		
Linear	1.13–1.33	1.23 (0.07)
Exponential	0.93–1.14	1.03 (0.07)
VEMAP	0.75–1.45	1.20 (0.25)
NH_4-N		
Concentration (mg/L)		
Linear	0.06–0.07	0.07 (0.01)
Exponential	0.04–0.05	0.04 (0.00)
Deposition (kg/ha)		
Linear	0.60–0.78	0.69 (0.06)
Exponential	0.38–0.54	0.45 (0.05)
VEMAP	0.32–0.65	0.44 (0.10)

Note: VEMAP = Vegetation/Ecosystem Modeling and Analysis Project ⟨http://www.cgd.ucar.edu/vemap⟩

1998, Interlandi and Kilham 1998). The experiments confirm that a slight N enrichment affects algal assemblages, but does not relate directly to atmospheric-deposition amounts. As with studies evaluating thresholds of organismal response to acidity and acid rain, experimental studies on organisms must be coupled with models of ecosystem biogeochemical dynamics (Baker and Christensen 1991). Biogeochemical models that identify and track N pools and fluxes through terrestrial ecosystems are necessary to relate atmospheric N deposition to soil and plant dynamics and leaching losses of N to surface waters. An ecosystem model that simulated stream responses to N deposition suggested alpine-stream N concentrations responded at 2.0 kg $N{\cdot}ha^{-1}{\cdot}yr^{-1}$, or slightly higher than hindcast estimates (Baron et al. 1994).

A paleolimnological reconstruction of diatom assemblages from four lakes in Wyoming revealed a shift in community structure similar to that observed in Colorado lakes (Saros et al. 2003). The onset of change began in the early 1990s. Atmospheric N deposition to the Beartooth Mountains in Wyoming had a range of $\leq$1.5 kg NO_3-$N{\cdot}ha^{-1}{\cdot}yr^{-1}$ for 1992–1999 (Nanus et al. 2003). Both NO_3-N and NH_4-N have increased significantly at Yellowstone National Park (WY08), the nearest NADP site, since 1985 (Nilles and Conley 2001). These data offer independent evidence that diatom community changes in alpine headwater lakes occurred in the range of 1.5 kg $N{\cdot}ha^{-1}{\cdot}yr^{-1}$. Directional changes in climate have also occurred in the Beartooth Mountains, however, complicating interpretation of why diatom changes occurred (Saros et al. 2003).

Difficulties arise from extrapolating deposition values backward in time. The lack of correspondence between dry and wet deposition values leads to de facto underestimates of total N deposition. Estimates based on the ROM406 CASTNET deposition-monitoring station suggest dry deposition adds 20–50% above wet deposition, but separate locations of CASTNET and NADP collectors hamper interpretations of total N deposition to Loch Vale. Dry N deposition was estimated at 40–50% of total wet inorganic-N deposition from co-located wet and dry collectors at Niwot Saddle (Sievering 2001). But the wet:dry ratio is not fixed, and will change as a result of changing emissions sources and nitrogen compounds. The wet deposition values used in this paper must therefore be viewed as minima, with no legitimate way to estimate a more realistic total N deposition.

There is a disappointingly poor relation between reported NO_x emissions and measured wet N or NO_3-N deposition. While some of this may be due to the few data pairs and uncertainty in reported emissions and deposition values, Butler et al. (2003) describe additional reasons for the difficulty in establishing quantifiable relations. Interannual variability in wet N deposition can be due to the inability of highly reactive NO_x to behave conservatively in the atmosphere. Since NH_4-N currently comprises nearly half of total inorganic wet N deposition, a poor correlation is to be expected. Mountainous terrain further influences site-specific deposition by creating turbulent winds and funneling emissions up one canyon but not another (Sievering 2001).

Even considering these caveats, experiments that show shifts in algal species assemblages, independent ecosystem model results, and Wyoming lake paleolimnological reconstructions support the hindcasting wet deposition estimate of 1.5 kg $N{\cdot}ha^{-1}{\cdot}yr^{-1}$ for the critical load. While there are complications from uncertainties in emissions, deposition, and confounding causes of environmental change, it appears hindcasting, particularly using equations that mimic population growth and NO_x emissions, is a reasonable method to derive a critical load if the dates for environmental change are known.

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