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ECOSYSTEMS

Ecosystem Responses to Nitrogen Deposition in the Colorado Front Range

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Abstract

We asked whether 3–5 kg N y^{-1} atmospheric N deposition was sufficient to have influenced natural, otherwise undisturbed, terrestrial and aquatic ecosystems of the Colorado Front Range by comparing ecosystem processes and properties east and west of the Continental Divide. The eastern side receives elevated N deposition from urban, agricultural, and industrial sources, compared with 1–2 kg N y^{-1} on the western side. Foliage of east side old-growth Englemann spruce forests have significantly lower C:N and lignin:N ratios and greater N:Mg and N:P ratios. Soil % N is higher, and C:N ratios lower in the east side stands, and potential net N mineralization rates are greater. Lake NO_3 concentrations are significantly higher in eastern lakes than western lakes. Two east side lakes studied paleolimnologically revealed rapid changes in diatom community composition and increased biovolumes and cell concentrations. The diatom flora is now representative of increased disturbance or eutrophication. Sediment nitrogen isotopic ratios have become progressively lighter over the past 50 years, coincident with the change in algal flora, possibly from an influx of isotopically light N volatilized from agricultural fields and feedlots. Seventy-five percent of the increased east side soil N pool can be accounted for by increased N deposition commensurate with human settlement. Nitrogen emissions from fixed, mobile, and agricultural sources have increased dramatically since approximately 1950 to the east of the Colorado Front Range, as they have in many parts of the world. Our findings indicate even slight increases in atmospheric deposition lead to measurable changes in ecosystem properties.

Key words: nitrogen; Rocky Mountains; Colorado; subalpine forests; alpine and subalpine lakes; paleolimnology; diatoms; N isotopes.

Introduction

The proportion of oxidized nitrogen and ammonia introduced to the atmosphere from human societal activities now exceeds that produced naturally (Vitousek and others 1997). Atmospheric deposition of N species has increased commensurately in some parts of the world, and ecological consequences include reduction in soil fertility, shifts in vegetation types, and lake and stream acidification (Stoddard 1994; Dise and Wright 1995; Vitousek and others 1997; Aber and others 1998). Most studies on ecological responses come from regions receiving chronically high N deposition loads, in excess of 10 kg N ha^{-1} y^{-1}. Additional insight comes from experimental manipulations, in which N is added to forests, soils, and streams at relatively high rates (Kahl and others 1993; Magill and others 1997; Emmett and others 1998). The eastern flanks of the Colorado Front Range have higher N deposition

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than most other areas of the state, but average wet plus dry N deposition is approximately 3–5 N ha^{-1} y^{-1}. More remote locations in Colorado report half as much wet N deposition (NADP 1999).

What are the consequences of low, chronic inputs of N? We explored this question in undisturbed Colorado Front Range ecosystems, looking for subtle evidence that changes due to N deposition have occurred. We emphasize the term subtle (compare McDonnell and Pickett 1993) and its application to ecological systems. McDonnell and Pickett (1993) state "Subtlety covers a wide variety of often inconspicuous or unexpected interactions of humans with ecosystems." With every human-mediated ecological change, there must be a point of initiation, in which ecosystem structure and function begin to respond. The alterations often are not detected until far down the trajectory of change. However, it is critical, albeit difficult, to detect them at earlier stages, when management or policy intervention can be applied most effectively.

In this article, we summarize ecological responses of high elevation ecosystems to N deposition. We and others have published observations and model results regarding chemical indications of excess N deposition in a few catchments (Lewis and Grant 1979, 1980; Lewis 1982; Baron 1992; Baron and others 1994; Campbell and others 1995; Williams and others 1996a). We have not addressed, however, whether there has been any biological response.

Our investigations in the Colorado Front Range looked for initial changes by examining: (a) forest and soil biogeochemical properties in regions of low and higher N deposition; (b) the spatial distribution of lakes influenced by enhanced N deposition; and (c) the response of algal diatom communities by using high-resolution paleolimnological records from alpine lakes. The prior literature documenting forest and soil responses to N deposition and the extensive documentation of diatom sensitivities to lake chemistry suggested to us these parameters would be most likely to detect change from N deposition if it had occurred. A survey of lakes adds weight to our suggestion of source areas. We took full advantage of the meteorological barrier that is the Continental Divide. Atmospheric deposition from sources east of the Divide move upslope to the highest elevations, but at this location air masses become entrained in high elevation winds and curve back to the east (Bossert 1990; Baron and Denning 1993). More remote mountain areas, unless exposed to point sources of NO_x emissions, have lower N deposition (NADP 1999).

Elevated nitrogen deposition has been identified as an environmental additive to higher elevations of the Colorado Front Range for years. Lewis and Grant (1980) found that a 3-year increase in precipitation acidity was significantly correlated with NO_3-N but not with SO_4 during the 1970s. Subsequent measurements documented the eastern side of the Front Range receives elevated wet N deposition, much of it associated with urban and agricultural air masses from the South Platte River basin to the east (Lewis and others 1984; Langford and Fehsenfeld 1992; Baron 1992; Williams and others 1996a; NADP 1999). Additional N is deposited in gaseous form, primarily during summer months (Parrish and others 1990; Langford and Fehsenfeld 1992; Sievering and others 1996; CASTnet 1999).

Much of the concern regarding N deposition to high mountains in Colorado stems from high and sustained NO_3 concentrations measured in east side alpine and subalpine lakes since 1982 (Baron 1983, 1992; Williams and others 1996a). However, other than high NO_3 concentrations, ranging between 12 and 40 μmol L^{-1} in alpine streams, and a mean annual value of 16 μmol L^{-1} in a nearby subalpine lake, there has been little additional chemical response (Baron 1992; Campbell and others 1995). For instance, springtime fluctuations in acid-neutralizing capacity (ANC) from 25 to 55 μmol L^{-1} in these alpine streams are not correlated with NO_3, but rather with dissolved organic carbon (DOC) and total aluminum (Al) (Denning and others 1991). This suggests soil organic acids, not strong acid anions from deposition, control ANC, and acidity, even during snowmelt.

Enhanced growth rates are often the first response for vegetation to additions of N (Bowman and others 1993; Baron and others 1994; Vitousek and others 1997; Aber and others 1998; Fenn and others 1998). Microbial nitrogen mineralization rates also respond rapidly to increasing N availability (McNulty and others 1991; Aber and others 1998), but long-term microbial activity responses to N are varied (Soderstrom and others 1983; Hunt and others 1988; Prescott 1995; Aber and others 1998). Climate, vegetation type, and stand age all influence the lag of ecological responses to increased N, but in general, mature vegetation in cool climates with short growing seasons will show symptoms of enhanced N availability sooner than earlier seral stages in more moderate climates (Fenn and others 1998; Magill and others 1997).

Excess N additions to aquatic ecosystems contribute to lake and stream acidification, although the European and North American regions reporting this acidification were concurrently influenced by sulfuric acid deposition (Schaefer and others 1990). In these regions, even though SO_4 concentrations have declined markedly from emissions reductions (Clow and Mast 1999), long-term depletion of soil base cations allows inputs of NO_3 to continue to reduce ANC and increase acidity (Likens and others 1996; Stoddard and others 1999). Even if acidification does not result, N additions can contribute to eutrophication if sufficient phosphorus is present. Reviews of nutrient limitation bioassays in North American lakes found N limitation to be about as common as P limitation (Elser and others 1990; Vitousek and others 1997). In a study of eight lakes in Colorado, Morris and Lewis (1988) found N to be most frequently limiting to phytoplankton growth. Long-term studies of Lake Tahoe have documented the slow shift of this large lake from N to P limitation as a result of atmospheric N deposition (Jassby and others 1994).

METHODS

Study Region: The Colorado Front Range

Surveys and research were conducted at elevations greater than 3000 m a.s.l. in forest and lake ecosystems of the Colorado Front Range (Figure 1). Additional forest and soil stands were located south and west of the Front Range. The Front Range is a north–south trending massif that defines the western edge of the large urban and agricultural South Platte River Basin. Bedrock composition in all forest stands is similar, and consists of Precambrian granite, schist, and gneiss (Lovering and Goddard 1959). Soils are shallow and coarse textured. Alpine soils are classified as Cryochrepts and Cryohemists, whereas forested soils are classified as Cryoboralfs (Baron 1992). Soils above tree line have pH values that range between 4.0 and 5.9, whereas forested soils are acidic, with pH values between 3.3 and 5.0 (Baron 1992; Stottlemyer and others 1997). Percent base saturation is highest in organic soil horizons; neutral salt base saturation is approximately 65% in Loch Vale forest soils in 1984, and 30% in underlying mineral soils. Organic soil base saturations in the Fraser Experimental Forest are reported as 55% by Stottlemyer and others (1997). Alpine tundra is found at elevations above approximately 3300 m, and Englemann spruce-subalpine fir forests occur below that (Peet 1988).

There are hundreds of lakes in the Front Range. We surveyed 44 for this study, on both east and west sides of the range. These lakes are clear, low ionic strength cirques that are ice covered from November to June.

The climate at high elevations is cold, with a mean annual temperature of 1.5°C in the forest at Loch Vale (Baron 1992), 1.3°C at Niwot Ridge (Williams and others 1996b), and 0.5°C at the Fraser Experimental Forest (Alexander and Watkins 1977). Mean annual precipitation is 1100 mm at Loch Vale (Baron 1992), 1200 mm at Niwot Ridge (Williams and others 1996b), and approximately 900 mm at Fraser Experimental Forest (Daly and others 1994). Most precipitation is deposited as snow between November and May.

Study Region: Characterization of Wet Atmospheric Deposition

Two high elevation wet deposition chemistry monitoring sites on the east side of the Front Range report mean N deposition of 3.6 and 3.5 kg ha^{-1} y^{-1} for Niwot Ridge and Loch Vale, respectively (Williams and others 1998; NADP 1999). Dry deposition measurements from a lower elevation (2850 m) monitoring site near Loch Vale suggest HNO_3, NO_3, and NH_4 aerosols contribute a minimum of 0.9 kg N ha^{-1} during April–September of each year (CASTnet 1999). There are no comparable monitoring sites west of the Continental Divide, but an independent measure of wet N deposition at Fraser Experimental Forest (3350 m) was 1.1 kg N ha^{-1} in 1990 (Stottlemyer and others 1997).

Because deposition reflects the amount of precipitation as well as the concentration of N compounds, it may mask actual differences in concentration due to different proximities to source areas. We compared NO_3 and NH_4 concentrations from all Colorado National Atmospheric Deposition Program monitoring sites to look at spatial pattern (Table 1; NADP 1999). Sites east of the Continental Divide had significantly higher concentrations of both NO_3 (1.48 mg L^{-1} east vs 1.08 mg L^{-1} west, $P = 0.01$) and NH_4 (0.37 mg L^{-1} east vs 0.15 mg L^{-1} west, $P = 0.00$).

Trend analyses of log-transformed NO_3 and NH_4 concentrations, by using a two-stage least squares general linear model (Lynch and others 1995), reveal there has been no trend in NO_3 concentrations at Sunlight Peak (-0.04 μmol L^{-1} y^{-1}) and Loch Vale (0.01 μmol L^{-1} y^{-1}) over time, but a significant increase of 0.09 μmol L^{-1} y^{-1} ($P = 0.01$) at Niwot Saddle. Significant ($P < 0.01$) increases in NH_4 concentrations were observed at both Loch Vale (0.18 μmol L^{-1} y^{-1}) and Niwot Saddle (0.26 μmol L^{-1} y^{-1}), but not at Sunlight Peak (-0.01 μmol L^{-1} y^{-1}).

Soil and Foliar Analyses

Twelve old-growth stands of Englemann spruce were selected for study, six east and six west of the Continental Divide on national forest and national

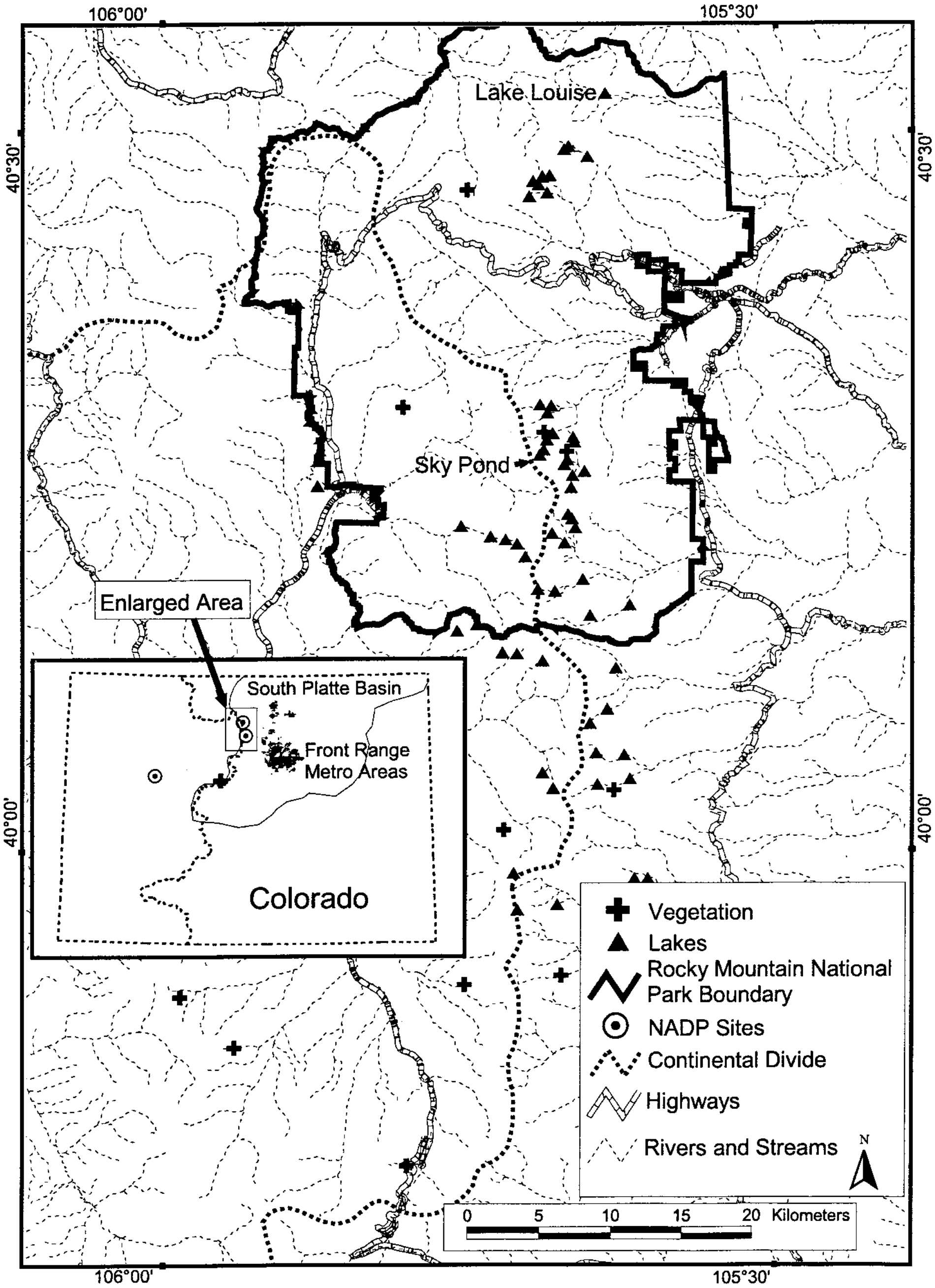

Figure 1. Map showing locations of vegetation (and soil) stands, lake sample sites, lakes cored for paleo records (Sky Pond and Lake Louise), and the three National Atmospheric Deposition Program (NADP) sites for which trend analyses were conducted. The west-most NADP site is Sunlight Peak; NADP sites east of Continental Divide are Loch Vale (north) and Niwot Saddle (south). Inset shows location of Front Range study area in Colorado, with the South Platte river basin outlined and major metropolitan areas identified.

park lands (Figure 1). We controlled for microsite climatic variability by selecting only northeast-facing stands at elevations between 3000 and 3500 m elevation. Current year foliage, and organic and mineral horizon samples were collected from three 30 × 30–m plots within each stand. Samples were analyzed for percent C and N, major base cations, P, and lignin. A 5-week laboratory soil incubation was performed to determine potential organic horizon net mineralization and nitrification rates. Statistical analyses using a mixed general linear model of analysis of variance compared east side with west side stands. Methods are described in detail in Rueth and Baron (unpublished).

Lake Chemistry Sampling and Analytical Methods

Samples were collected from the surface or outlets of 19 lakes in 1997 and from 14 lakes in 1998. An additional 11 lakes sampled in 1995 by Musselman and others (1996) were added to increase the sample size for lakes west of the Continental Divide. Sampling techniques, including those from Musselman and others (1996), were similar for all lakes

Table 1. Volume-Weighted Mean (VWM) Weekly Concentrations of NO_3 and NH_4 (μmol L^{-1}) in Wet Atmospheric Deposition at All 17 NADP Sites in Colorado

	Latitude and Longitude	Elevation (m)	No. Weeks Analyzed	Mean Annual Precipitation (mm) (SE)	VWM NH_4 (μmol L^{-1}) (SE)	VWM NO_3 (μmol L^{-1}) (SE)
East side						
CO00 Alamosa	37°26′29″ 105°51′55″	2298	517	188 (11)	19.09 (0.93)	17.55 (0.79)
CO01 Las Animas	38°07′04″ 103°18′58″	1213	459	350 (17)	29.05 (1.17)	29.35 (1.11)
CO02 Niwot Saddle	40°03′19″ 105°35′18″	3520	568	1200 (132)	16.68 (2.55)	24.55 (1.83)
CO19 Beaver Mdws	40°21′51″ 105°34′55″	2490	705	390 (21)	14.77 (0.61)	21.40 (0.68)
CO21 Manitou	39°06′04″ 105°05′31″	2362	701	400 (15)	14.19 (0.61)	26.81 (0.96)
CO22 Pawnee	40°48′23″ 104°45′17″	1641	614	340 (16)	41.51 (2.60)	32.62 (1.07)
CO94 Sugarloaf	39°59′38″ 105°28′48″	2524	481	543 (22)	19.51 (0.85)	24.01 (0.78)
CO98 Loch Vale	40°17′16″ 105°39′46″	3159	622	1100 (46)	9.46 (0.69)	15.14 (0.51)
West side						
CO08 Four Mile Park	39°24′11″ 107°20′28″	2502	430	544 (37)	7.61 (0.52)	14.62 (0.64)
CO15 Sand Spring	40°30′27″ 107°42′07″	1998	745	345 (18)	12.42 (0.72)	23.63 (0.90)
CO91 Wolf Creek Pass	37°38′07″ 106°47′25″	3292	265	150 (24)	5.73 (0.29)	12.86 (0.51)
CO92 Sunlight Peak	38°25′38″ 107°22′47″	3206	395	680 (32)	8.28 (0.46)	16.28 (1.80)
CO93 Dry Lake	40°32′05″ 106°46′48″	2527	487	800 (47)	9.30 (0.49)	20.49 (0.85)
CO95 Engineer Mtn.	37°39′35″ 107°47′57″	2758	94	679 (110)	7.15 (1.35)	17.87 (3.13)
CO96 Molas Pass	37°45′05″ 107°41′07″	3249	419	770 (36)	5.57 (0.30)	13.81 (0.53)
CO97 Buffalo Pass	40°32′16″ 106°40′35″	3234	598	1040 (75)	7.22 (0.31)	14.12 (0.52)
CO99 Mesa Verde NP	37°11′53″ 108°29′25″	2172	605	48 (22)	10.00 (0.57)	23.09 (0.90)
East side average					20.53	23.93
West side average					8.14	17.42

Values were summarized from the NADP data base (1999). The east and west side averages differ significantly with a one-tailed t test ($P < 0.01$). Standard error = (SE).

(EPA 1987). Most samples, including those from Musselman and others (1995), were analyzed by the USDA Forest Service Rocky Mountain Station Biogeochemistry Laboratory (EPA 1987). Ten percent of all samples were collected in duplicate for quality control, and they agreed within 5%.

Long-term sample collections at The Loch, an east side lake in Rocky Mountain National Park, reveal considerable seasonal and interannual variability in NO_3 during the months of July through September for years 1982–98 (Baron 1992). This variability, up to 8 μmol NO_3 L^{-1} between years or even between late summer months, precludes simple statistical comparisons of the 50 lakes sampled on different days in 1995, 1997, and 1998. The survey lake concentrations therefore were adjusted, or normalized, with respect to lake concentrations from The Loch to minimize the effects of yearly and seasonal variability. Although the variability in late summer conductivity and ANC is not nearly as pronounced as for NO_3, these values also were adjusted against the long-term Loch records. We assumed solute concentration response over time was similar in the survey lakes to that of The Loch, an assumption borne out by a comparison of temporal coherency of two catchments with long-term records (Baron and Caine 2000). A smoothing spline was used to obtain curves that adequately fit the scatter plots for the Loch data from July and August 1995, 1997, and 1998 (Hastie and Tibshirani 1990). Fitted values from The Loch curves for corresponding lake survey sample dates were obtained by simple linear interpolation of the predicted values from the smoothing spline.

The adjusted solute value for each survey lake was produced by subtracting the corresponding Loch value from the lake's solute concentration. Analysis of variance was used to test for differences between adjusted solute concentrations of east and west side lakes over the years studied. No violations of the ANOVA assumptions were observed.

Sediment Collection Methods, Analysis for Diatoms, and N Isotopes

Gravity cores of sediments from Sky Pond and Lake Louise, two alpine lakes east of the Continental Divide, were collected in 1997 and extruded in consecutive 0.5-cm (upper 10.0 cm) or 1.0-cm (below 10.0 cm) increments in the field (Figure 1; Wolfe and others unpublished; Glew 1988, 1989).

The chronology of recent sediments was based on α-spectroscopic measurements of sediment ^{210}Pb activity, to which the constant rate of supply (CRS) model was applied (Appleby and Oldfield 1978). Sediment $\delta^{15}N$ was determined by isotopic ratio mass spectrometry of combusted and cryogenically purified samples (Fry and others 1992). Measurements were reproducible within ±0.1%. Diatom slides for light microscopy were prepared from digested (30% H_2O_2) sediment samples by using standard techniques (Battarbee 1986). At least 500 diatom valves were enumerated from each sample under oil immersion at ×1000 magnification, along random transects including cover slip edges. Diatom taxonomic identifications followed standard floras (for example, Patrick and Reimer 1966, 1975; Foged 1981; Germain 1981; Krammer and Lange-Bertalot 1986–91). Diatom valve concentrations were established by the addition of known quantity of exotic markers, in this case *Eucalyptus* pollen grains from a calibrated slurry (Wolfe 1997). Diatom assemblages are expressed as relative frequencies of individual or grouped taxa in relation to the sum of total diatom valves counted. With the objective of producing an index of diatom paleoproductivity, diatom valve concentrations also were converted to total diatom biovolume, by using a combination of measured and published average cell volume estimates for each of the taxa encountered (Battarbee 1973; Anderson 1994). Diatom floristic changes were evaluated objectively by rate-of-change analysis (Jacobson and Grimm 1986), which is the calculation of a between-sample multivariate dissimilarity metric (in this case squared chord distance), divided by the time elapsed between the deposition of subsequent samples. Rate of change results are only indicated for the upper 10 cm of the cores, which are well constrained by the ^{210}Pb chronologies.

Results

Forest Foliage and Soils

East side forest stands had significantly greater percent foliar N (1.09% N) than west side stands (0.96% N; Table 2). East side stands had lower foliar C:N. Foliar lignin content was not significantly different between east and west side stands, although the lignin:N ratio was lower at east side stands. Magnesium was significantly depleted, and P was significantly enriched in foliage of east side stands. Nitrogen:magnesium, N:Ca, and N:P ratios were significantly greater in east side stands (Table 2).

Table 2. Foliar and Soil Chemistry and Soil Microbial Responses from Six East and Six West Side Forest Stands of the Colorado Front Range

	East Side	West Side
Foliar chemistry		
% N	1.09 (0.08)	0.96 (0.1)[a]
C:N	47.30 (3.6)	53.10 (6.3)[a]
% Mg	0.093 (0.01)	0.10 (0.01)[b]
% P	0.18 (0.03)	0.25 (0.03)[b]
N:Mg	11.80 (1.60)	9.49 (1.50)[b]
N:P	6.21 (0.89)	4.76 (0.75)[b]
Ca:Al	59.6 (33.4)	60.8 (30.3)
% Lignin	15.82 (0.7)	16.16 (1.2)
Lignin:N	14.63 (1.2)	17.01 (2.2)[b]
Soil characteristics		
Organic soil % N	1.39 (0.2)	1.08 (0.2)[b]
Organic Soil % C	35.8 (4.8)	34.2 (5.9)
Organic soil C:N	25.9 (2.7)	32.5 (5.0)[b]
% Lignin	30.5 (4.2)	29.6 (5.8)
Lignin:N	22.2 (3.1)	28.3 (5.7)[b]
Microbial mineralization rate	3.42 (2.7)	0.69 (1.0)[b]
Microbial nitrification rate	0.57 (1.5)	0.06 (0.3)

Values are means and standard deviations (in parentheses). Mineralization rates are in $\mu g\ N\ g^{-1}\ d^{-1}$. There were six degrees of freedom for foliar samples and 10 degrees of freedom for soil samples.
[a]Significance at 0.1 level.
[b]Significance at 0.05 level (from Rueth and Baron unpublished).

Soils underlying the old-growth forest stands of west and east sides had similar soil properties. There was significantly greater organic soil % N and significantly lower C:N ratios in east side soils versus west side soils (Table 2). Percent C did not differ between east and west sides, averaging 35%. Organic soil percent lignin was not significantly different between sides, but the lignin:N was significantly lower at east side stands. The average soil N pool, calculated from the average % N and average soil bulk density (6935 g soil m^{-2}), was 965 kg N ha^{-1} for east side stands and 750 kg N ha^{-1} for west side stands.

Potential net mineralization rates were significantly greater in east side soils than west side soils (Table 2). Potential net nitrification rates were low for most stands. Mineralization rates increased linearly with organic soil with greater than 1.2% N (Figure 2). West side soils had organic soil with less than 1.4% N, and low mineralization rates. East side soils had greater than or equal to 1.3% N and high mineralization rates (Figure 2).

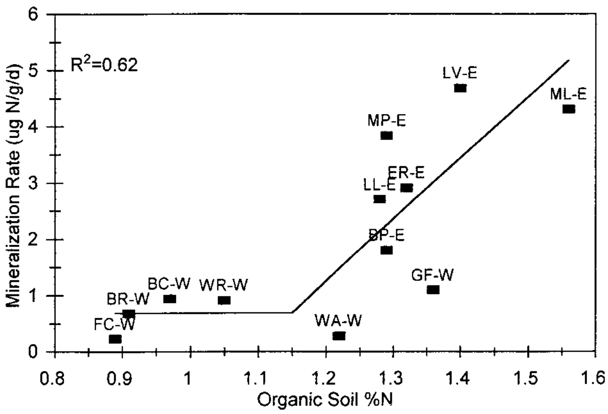

Figure 2. N mineralization rate compared with organic soil % N in east side forest soils (sites with a -E suffix) and west side forest soils (sites with a -W suffix). Data from (Rueth and Baron unpublished).

Lake Chemical Surveys

All lakes were dilute, with specific conductivity ranging from 5.8 to 21.1 microSiemens (μS) cm^{-1} and a median value of 8.6 $\mu S\ cm^{-1}$ (Table 3). ANC ranged from 23.6 to 134.7 $\mu mol\ L^{-1}$, with a median of 46.8 $\mu mol\ L^{-1}$, whereas pH ranged from 6.0 to 7.0, with a median value of 6.4. Phosphorus was measured as PO_4 in most lakes but was at or below the analytical detection limit of 0.021 $\mu mol\ L^{-1}$. Nitrate in all lakes ranged from below detection limits to 20.2 $\mu mol\ L^{-1}$, whereas NH_4 was mostly undetectable but ranged as high as 4.8 $\mu mol\ L^{-1}$. The median values for NO_3 and NH_4 were 8.5 and less than 0.05 $\mu mol\ L^{-1}$, respectively.

The mean nitrate concentration of the east side lakes (10.5 $\mu mol\ L^{-1}$) was significantly higher than the west side lakes (6.6 $\mu mol\ L^{-1}$; $P = 0.02$ for adjusted values). The location of the lakes on east or west sides was significant in explaining variation in the adjusted nitrate concentrations ($P = 0.03$). Neither the year of sample collection nor the year coupled with location influenced adjusted NO_3 concentrations. There was no pattern of concentration for conductivity with lake location east or west (8.8 compared with 10.3, $P = 0.12$), but ANC was significantly lower in east side lakes, with a mean of 49.0 compared with the west side mean of 72.2 ($P = 0.01$ for adjusted values).

Lake Sediment Records

Before 1900, diatom assemblages in both Sky Pond and Lake Louise were dominated by *Aulacoseira* spp., *Fragilaria pinnata*, *F. construens* var. *venter*, and *Achnanthes* spp., a typical oligotrophic alpine flora (Figure 3). Sometime before approximately 1950, the mesotrophic planktonic taxa *Asterionella formosa* and *Fragilaria crotonensis* became common elements of diatom assemblages, and between approximately 1950 and approximately 1970 *A. formosa* became the dominant diatom in both lakes. Both taxa are present in trace frequencies in older sediments, implying their increases were environmentally stimulated, rather than the result of colonization (Wolfe and others unpublished). Valve concentrations and diatom biovolumes increased dramatically after 1950, and their rates of change since mid-20th century are greater than those that occurred since the 1700s (Figure 4). Ecological rates of change since 1950 are greater than the last several hundred years, and actually greater than over the entire 14,000-year record of natural edaphic, climatic, or limnological processes (data not shown).

The sediment $\delta^{15}N$ measurements show that values were stable between 4 and 5‰ in Sky Pond and Lake Louise before approximately 1900 but began to decrease shortly afterwards, by approximately 1‰ in the first half of the century (Figure 4A). The trend towards lighter isotopic values accelerated after approximately 1950 for Sky Pond, and after approximately 1970 for Lake Louise, with further depletions of 1.0 and 2.5‰, respectively (Wolfe and others unpublished). The changes in $\delta^{15}N$ are nearly coincident with the changes in diatom flora; neither algae nor $\delta^{15}N$ correspond with other signals of anthropogenic influence recorded in the lake sediments over this period, such as increases in atmospherically deposited lead (Wolfe and others unpublished).

Discussion

Cumulative Evidence

A summary of results show that N deposition is greater on the east side of the Front Range, east side forests and soils demonstrate biogeochemical traits consistent with elevated N deposition, and east side soils additionally have greater rates of microbial N mineralization than west side sites (Table 4). East-side lakes have higher NO_3 concentrations, and at least some of them reflect an algal community flora indicative of nutrient enrichment and disturbance. Current N isotopic ratios are unique in the 14,000-year history of alpine lakes. We think the evidence is strong that excess N deposition has caused the observed responses. But are there plausible alternative explanations for our observations? We explore these below.

Table 3. Characteristics for High Elevation Lakes Sampled from East (E) or West (W) of the Continental Divide in the Colorado Front Range

Lake Name	East/ West	Date (yymmdd)	Elevation (m)	Cond. (μS cm^{-1})	pH	ANC (μmol L^{-1})	NO_3 (μmol L^{-1})	NH_4 (μmol L^{-1})
Rainbow #2	E	980805	3109	21.1	6.66	134.7	9.1	<0.5
U. Coney	E	980807	3334	13.2	6.60	72.7	9.2	<0.5
Coney	E	980808	3230	20.2	6.73	117.7	11.9	<0.5
Mitchell	E	980810	3267	13.5	6.42	68.1	6.8	<0.5
Blue	E	980810	3450	6.8	6.29	24.7	8.4	<0.5
Falcon	E	980812	3371	7.1	6.23	38.1	6.5	<0.5
Snowbank	E	980813	3511	8.9	6.59	46.8	9.5	<0.5
Lion #1	E	980813	3372	10.5	6.60	58.4	8.8	<0.5
Lion #2	E	980813	3474	8.3	6.58	44.9	8.6	<0.5
Thunder	E	980814	3222	9.3	6.57	61.7	8.3	<0.5
Pipit	E	980817	3479	6.7	6.53	36.3	7.2	<0.5
Bluebird	E	980817	3346	8.9	6.39	46.4	9.3	<0.5
Isabelle	E	980829	3312	11.2	6.64	46.7	5.9	2.93
Long	E	980829	3206	14	6.40	87.4	1.9	4.81
Red Deer	E	950807	3161	12.6	6.36	76.7	3.6	0.67
Emerald	E	970701	3072	10.5	6.35	47.1	19.3	2.03
Frozen	E	970702	3529	7.0	−9	30	18.5	<0.5
Black	E	070703	3236	6.9	6.16	25.1	15.6	<0.5
Solitude	E	970704	3480	7.9	6.18	30.8	16.1	<0.5
Shelf	E	970704	3419	7.8	6.27	33.5	14.9	<0.5
Jewel	E	970704	3032	7.2	6.34	34.2	10.1	<0.5
Mills	E	970704	3029	7.6	6.39	36.3	10.8	<0.5
Lit. Crystal	E	970815	3508	8.6	6.19	57.7	1.0	<0.5
Crystal	E	970816	3511	9.1	6.42	43.8	7.4	<0.5
Lawn	E	970816	3288	13.9	6.53	91.1	8.7	<0.5
Upper Fay	E	970804	3413	7.9	6.26	27.5	20.1	<0.5
Middle Fay	E	970804	3276	8.0	6.41	40.3	13.2	<0.5
U. Spectacle	E	970804	3462	5.8	6.22	23.6	12.5	<0.5
Chiquita	E	970804	3459	7.6	6.39	36.2	8.5	<0.5
Dream	E	970730	3017	8.5	6.44	46.8	13.5	<0.5
Columbine	W	950731	3364	14.7	7.02	112.4	7.8	0.28
Crater	W	950807	3133	8.5	6.11	51.7	4.1	0.39
Dorothy	W	960807	3679	6.4	6.41	32.5	6.7	1.26
Island	W	950811	3474	15.4	6.44	119.5	8.5	<0.5
Round	W	950803	3401	6.9	5.96	41.5	6.6	0.39
Stone	W	950807	3243	10.4	6.35	73.6	3.1	0.39
Triangle	W	950812	3389	12.8	6.74	70.9	16.3	0.09
Upper	W	950808	3270	14.7	6.38	87.9	4.8	0.39
Watanga	W	950810	3288	13.0	6.51	128.4	<0.5	<0.5
Spirit	W	970706	3136	7.9	6.47	53.3	4.9	<0.5
5th	W	970707	3307	8.1	6.36	45.9	14.2	<0.5
4th	W	970707	3163	7.6	6.37	50	6.7	<0.5
Verna	W	970707	3108	9.2	6.56	71.2	5.2	<0.5
Lone Pine	W	970708	3012	9.0	6.45	71.8	4.0	<0.5
Mean east (SD)				10.0 (3.8)	6.40 (0.2)	52.9 (27.2)	10.5 (5.0)	<0.5 (1.2)
Mean west (SD)				10.3 (3.2)	6.40 (0.3)	72.2 (30.1)	6.6 (4.3)	<0.5 (<0.5)

All lakes were located either in Rocky Mountain National Park or in the Indian Peaks Wilderness.

Forest Ecosystem Processes

Alternative hypotheses. Forest ecosystem processes are guided and constrained by species composition, forest maturity, level of disturbance (such as fire, insects, or disease), climate, and nutrient and water availability. We minimized the influence

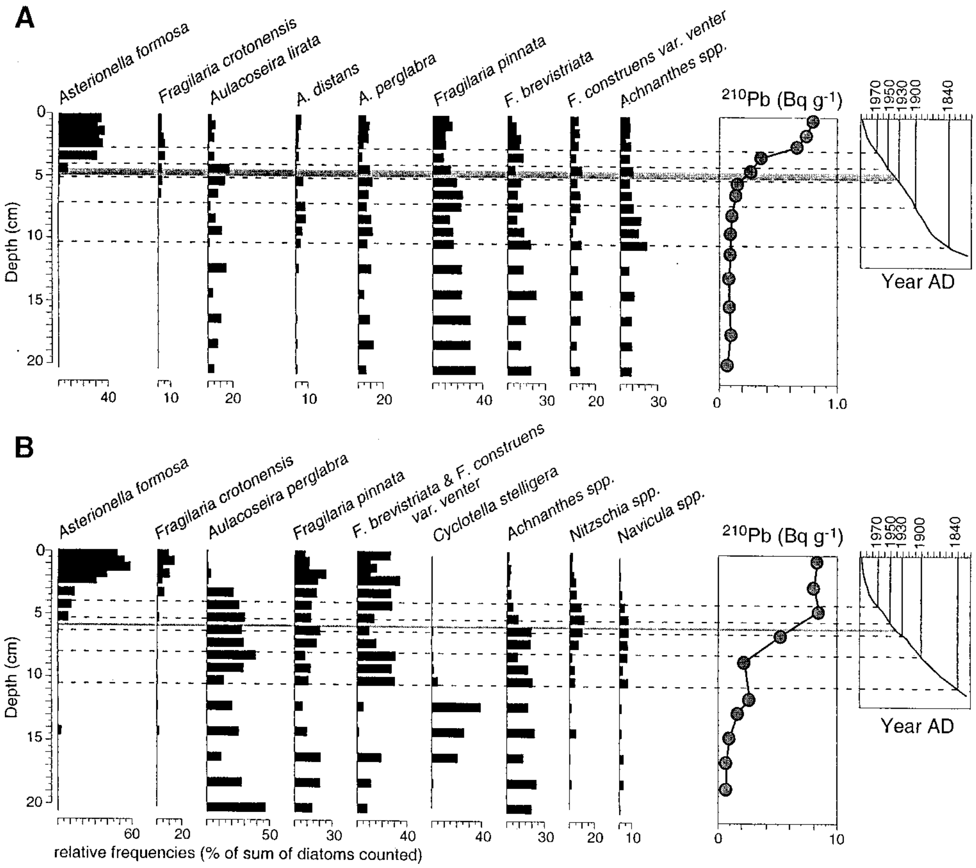

Figure 3. Relative frequencies of dominant diatom taxa, stratigraphic profiles of sediment ^{210}Pb activity, and age–depth relationships based on the CRS models for the gravity cores from (**A**) Sky Pond and (**B**) Lake Louise. The horizontal gray lines indicate fish introductions to the lakes in the 1930s.

of as many processes as possible by selecting >250-year-old Englemann spruce stands with a *Vaccinium* spp. understory. Subalpine fir and lodgepole pine were present in some plots, but always as a minor overstory component. Fire in subalpine forests recurs at 200- to 400-year intervals (Peet 1988), so these forests do not appear to be stressed by logging or burning practices common around the turn of the 20th century or by subsequent fire suppression activities (Veblen and Lorenz 1991). Similarly, stands showed no evidence of insect or disease outbreaks.

Soil physical characteristics and bedrock parent material were similar across sites (Rueth and Baron, unpublished). Mineral soil percent clay was significantly greater at east side sites, but the absolute difference between east side and west side soils was only 4.9%. Nevertheless, increased clay content directly influences water retention capability, perhaps allowing east side forest stands to store and cycle N more effectively.

Was there enough atmospheric deposition of N to account for the observed difference of 215 kg N ha^{-1} between east and west side sites? Human population in the South Platte Basin has risen exponentially since 1900 (Figure 5). Assuming N deposition increased commensurate with population numbers, we fit an exponential curve to N deposition rates that passed through a window of 4.7 kg N ha^{-1} in 1995. We also assumed 95% retention of N in soil organic matter, a figure suggested by simulations of subalpine forests with the CENTURY model (Baron and others 1994). If deposition paralleled population growth, it accounts for 162 kg N ha^{-1}, or 75% of the observed difference in soil organic matter N pools (Rueth and Baron, unpublished). Sixty percent of the N loading to soils would have occurred after 1950. There is obvious uncertainty in these numbers, but they plausibly suggest deposition could account for much of the observed differences in soil N pools.

We controlled for microclimate during site selection by choosing sites on northeast-facing slopes, between 3000-m elevation and the alpine treeline. There still could be mesoscale climatic differences in precipitation and temperature; the climate records are insufficient to directly address this question. An ecosystem simulation of a catchment in the study region that varied annual and seasonal precipitation and temperature by ±10% and ±2°C, respectively,

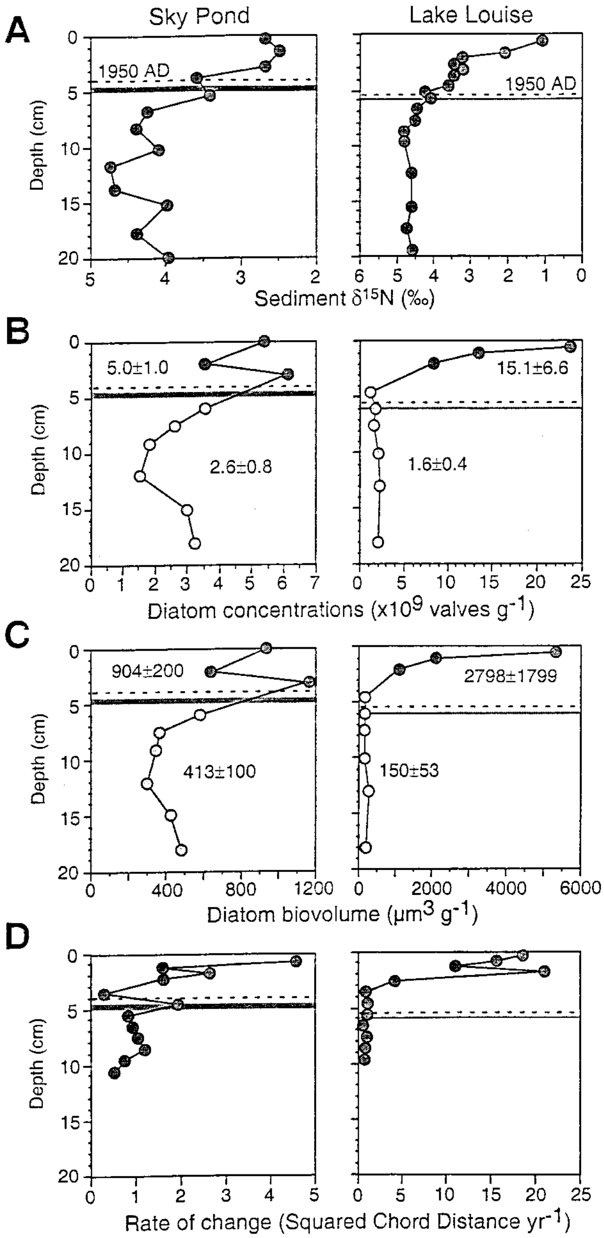

Figure 4. (**A**) Sediment δ^{15}N, (**B**) diatom valve concentrations, (**C**) diatom biovolumes, and (**D**) stratigraphic rates of change from the gravity cores collected at Sky Pond (left panels) and Lake Louise (right panels). Horizontal dashed lines indicate 1950 AD, according to the ^{210}Pb chronologies of each core. Numbers in (**B**) and (**C**) are mean values ± 1 standard deviation for the uppermost (dark grey circles) and lower (light grey) samples, reflecting the post- and predisturbance intervals of each record, respectively. The horizontal gray lines indicate fish introductions to the lakes in the 1930s.

showed no forest productivity response to precipitation and a 10% increase or decrease in net annual photosynthesis to warming or cooling (Baron and others 2000). These results suggest that climate variability could account for some of the observed differences in N cycling rates observed between east and west side stands. Warmer temperatures, however, are expected to widen the C:N ratio in soils and foliage, whereas, in fact, our warmer, east side stands had lower C:N ratios than the cooler, west side stands (Rueth and Baron, unpublished).

Similarities to other studies. Other studies have reported similar results to ours along spatial gradients of increasing N deposition. McNulty and others (1991) measured a linear increase in forest floor percent N in high elevation spruce-fir forests of the northeast US. Forest floor percent N increased from 1.09% at (low) wet N deposition values of 2.9 kg N ha^{-1} y^{-1}, up to approximately 2.16% N at (high) wet N depositions of 5.1 kg N ha^{-1} y^{-1} (McNulty and others 1991). Tietema and Beier (1995) showed a nearly identical linear relation between N deposition, forest floor % N, and foliar % N in coniferous forests across Europe, where deposition ranged from 2.7 kg N ha^{-1} y^{-1} at Sogndal, Norway, to 33 kg N ha^{-1} y^{-1} at Ysselsteyn in the Netherlands. These investigators, as well as others (Friedland and others 1988; Schulze and others 1989), found significant correlations between N deposition and foliar N:Mg ratios. West side stand forest floor C:N ratios of 32.5, and east side stand forest floor ratios of 25.9 are within the range of C:N values reported by McNulty and others (1991). High elevation old-growth coniferous forests from the Integrated Forest Study reveal a relation between N deposition and forest floor C:N ratios similar to our findings for east side and west side stands (Johnson and Lindberg 1992). Our west side C:N ratio is similar to the C:N ratio of 33 reported for the Findlay Lake site in the Cascade Mountains, a relatively unpolluted old-growth Pacific silver fir stand where deposition is estimated at 2.0 kg N ha^{-1} y^{-1}. Our Colorado east side stand C:N ratio is closer to that reported for Whiteface Mountain in New York (C:N of 20), where N deposition is reported at 15.9 kg N ha^{-1} y^{-1} (Johnson and Lindberg 1992; Fenn and others 1998).

N mineralization rates were higher in the east side stands of our study than the west side stands (Figure 2; Table 2). The observed low potential organic soil nitrification rates at most of our sites could be related to C:N ratios. A nitrification threshold has been postulated to exist where nitrification is limited in soils with a C:N ratio above approximately 24 (Gundersen and Rasmussen 1990; Emmett and others 1998). Our east side stands, at C:N of 25.9, are just above this threshold.

The plot of mineralization with % N of the forest

Table 4. Summary of Observed Phenomena Related to N Excess in the Colorado Front Range and Vicinity

Observations	East Side	West Side
Wet atmospheric deposition	Higher	Lower
Spruce foliar chemistry		
C:N	Lower	Higher
N:Mg	Higher	Lower
N:P	Higher	Lower
Lignin:N	Lower	Higher
Soil characteristics		
% N	Higher	Lower
C:N	Lower	Higher
Microbial characteristics		
Mineralization	Higher	Lower
Lake NO_3 concentration	Higher	Lower
Lake sediment diatoms		
Species composition	Representative of enrichment/disturbance	Not available
Biovolume	Greater than before 1950	Not available
Lake sediment N isotopes	Lighter than before 1950	Not available

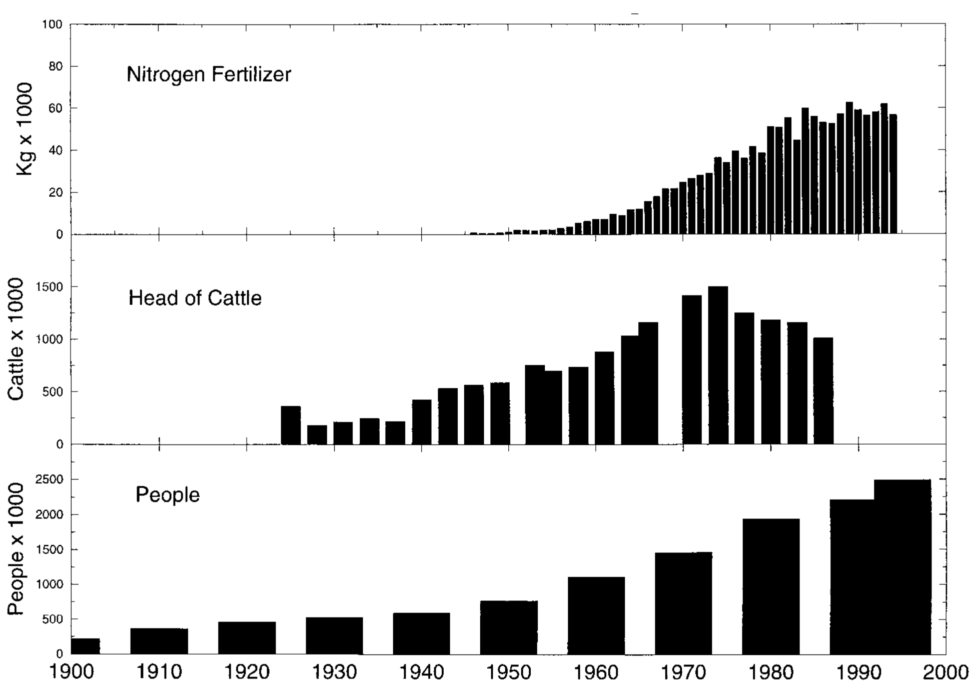

Figure 5. Trends in source of nitrogen emissions for 10 counties of the South Platte River Basin, Colorado (Adams, Arapaho, Boulder, Douglas, Jefferson, Larimer, Logan, Morgan, Washington, and Weld counties).

floor is remarkably similar to that presented by McNulty and others (1991) for nitrification rates with % N in forest floor (Figure 2). McNulty and others (1991) suggest a threshold of 1.4 % N above which nitrification increases linearly. The increase in mineralization rates in the Colorado soils occurs at greater than 1.1% N. Aber and others (1998) report increased N mineralization rates in five studies of either N additions or transects across depositional gradients at early stages of fertilization or midgradient. Longer-term responses to N additions caused mineralization rates to decline, however. Similar results were reported by Tietema (1998) and Gundersen and others (1998).

In addition to high net nitrification rates in soils, there are three other biogeochemical indicators of N excess reported in the literature that we did not observe (Aber and others 1998; Gundersen and others 1998). These were decreased foliar Ca:Al, decreased foliar lignin concentrations, and elevated nitrification rates. These observations are consistent with placement of Colorado Front Range ecosystems early in the progression of symptoms related to N excess.

Lake Chemical Composition

Alternative hypotheses. Atmospheric deposition is only one of many factors that influence lake water chemical composition. The type and amount of upstream vegetation, lake area and volume, lake trophic status, and hydrologic flushing rates all can influence lake N concentrations (Baron and Campbell 1997). By selecting only lakes greater than 3000 m a.s.l. elevation, and eliminating any with extensive wetlands, we controlled somewhat for vegetation type. Lakes in this study were located in headwater basins with either talus, tundra, or subalpine forest vegetation upstream. All lakes were less than 10 ha and were glacial in origin.

Conductivity was not different between east and west side lakes, attributing to their similarity. Alkalinity was significantly lower on the east side, but this is not inconsistent with elevated N deposition. The net effect of NH_4 oxidation and NO_3 leaching through soils is acidification, so the loss of ANC could portray a trend toward acidification (Reuss and Johnson 1986).

Similarities to other studies. Our sample lakes were similar to (and some overlapped) those sampled in the Southern Rockies region as part of the Western Lake Survey in 1985 (Eilers and others 1987; Baker and others 1990). The median Southern Rockies lake area was 3.7 ha, median depth was 5.2 m, residence time was 0.29 years, and the watershed:lake area ratio was 24 (Baker and others 1990). The median NO_3 concentration for the 132 Southern Rockies lakes was 0.5 μmol L^{-1}. East side lakes of the Front Range sampled as part of the Western Lake Survey (WLS) had a median of 5.8 μmol L^{-1}, with a range of 0.1–15.5 μmol L^{-1} (n = 33 for lakes greater than 3000 m). West side lakes of the Front Range had a median of 0.9, with a range of 0.1–5.6 (n = 11 for lakes greater than 3000 m). Our findings, then, are consistent with those reported more than a decade ago showing east side NO_3 concentrations greater by more than a magnitude than west side concentrations.

Lake Diatom Community Composition

Alternative hypotheses. It is clear the diatom flora of these lakes has changed, but is excess nitrogen deposition the cause? A study of nutrient limitations to eight large lakes in Colorado found west side lakes to be N limited through most of the summer, but east side lakes were either P limited or colimited by N + P (Morris and Lewis 1984). Other studies of nutrient-poor lake districts also have shown primary productivity in many lakes to be N limited, a situation that enhances biotic sensitivity towards atmospheric N deposition (Elser and others 1990; Axler and others 1994; Jassby and others 1994). However, two other disturbances potentially could be responsible for a shift in algal communities and an increase in biomass: introduction of nonnative fishes and episodic transport of phosphorus to alpine lakes.

Most of the lakes in the Colorado Front Range were originally without fish but were stocked with trout beginning around the turn of the century (Rosenlund and Stevens 1990). Sky Pond was stocked from 1931 until 1939 and maintains a reproducing population of brook trout. Lake Louise was stocked once in 1934, but no fish were found in 1958 and 1970 surveys. Fish were observed in Lake Louise in 1995, but their populations are unknown, and apparently sporadic. Five to seven thousand fish were added with each stocking; in Sky Pond this was repeated at least five times over the years. Fish can alter the trophic status of lakes by consuming large invertebrate herbivores that graze on phytoplankton (Carpenter and others 1985). In oligotrophic lakes, fish also may reintroduce a significant amount of N and P from the sediments as body excretions (Brabrand and others 1990; Leavitt and others 1994). Both reduction of grazers and addition of nutrients from excreta should stimulate algal response. In Canadian alpine lakes, algal responses were commensurate with stocking (Leavitt and others 1994). However, the Colorado lakes were stocked in the 1930s, with some algal response, but the greatest change in *A. formosa* was not observed until approximately the 1950s for Sky Pond, and approximately the 1970s for Lake Louise (Figures 3 and 4). The offset in disturbance and response suggests fish introductions alone cannot explain the dramatic change in lake algal communities.

Other investigators have shown that atmospheric deposition can be an important source of P to lakes (Cole and others 1990; Jassby and others 1994), although both these groups found P to originate from a local, not regional, source area. Phosphorus has no gaseous component, and the flux of P through the atmosphere as soil dust is quite small compared with other pathways, such as nutrient cycling and hydrologic transport (Schlesinger 1997). Nevertheless, one could postulate episodic high-wind transport of P from the fertilized croplands on the plains to the mountains, where cycling within lake ecosystems would allow a long-term boost in primary productivity. Episodic inputs also could explain the significantly higher foliar % P found in east side forest stands.

There are no good records of atmospherically deposited P, because soluble PO_4 is rarely above detection limits (NADP 1999), and total P is not measured. However, bioassays conducted in alpine ponds in 1999 showed a positive response in productivity when P or N + P were added, suggesting algae are still P limited (K.R. Nydick and B. Moraska, unpublished data).

Similarities to other studies. *Asterionella formosa* is now the dominant diatom preserved in the sediment record from the two lakes we have cored, and *Fragillaria crotonensis* the subdominant diatom. These are opportunistic algae that respond rapidly to disturbance and slight nutrient enrichment in many parts of the world. They are among the first diatoms to follow catchment settlement and agriculture in European lakes in the 12th and 13th centuries (Anderson and others 1995; Lotter 1998), and North American settlements in the 18th and 19th centuries (Christie and Smol 1993; Hall and others 1999). In these studies, as well as in a lake in Sweden influenced by acidic deposition, the two species expanded after initial disturbance and were later replaced by other species more tolerant of either acidification or eutrophication (Renberg and others 1993; Hall and others 1999). Moreover, the growth of *A. formosa* was stimulated with N amendments [both $Ca(NO_3)_2$ and HNO_3] in in situ incubations in an east side lake in Rocky Mountain National Park (McKnight and others 1990).

N Isotopic Signatures

Alternative hypotheses. The distinctively light isotopic signatures in recent lake sediments could be due to four separate, nonmutually exclusive agents of change: a change in terrestrial watershed processing of N that would favor different N cycling pathways; a shift in algal communities with different fractionation efficiencies with respect to available N; change in internal lake N cycling, perhaps due to fish introductions; or a shift in the emission sources that provide N to the Front Range as atmospheric deposition.

The catchments above Sky Pond and Lake Louise are primarily talus and glacier, with slight amounts of alpine tundra. These conditions have been stable for the past several hundred years (Menounos and Reasoner 1997), so there is no reason to expect large changes in terrestrial microbial activity that could lead to such a large variation in sediment N isotope values.

Fish introductions influence lake N cycling by consuming sediment dwellers, zooplankters, and insects and excreting fecal material. But fish tissues and solid waste are enriched relative to diet from excretion of light isotopic N in urine (Kendall 1998). Further enrichment of waste comes from volatilization of ^{15}N-depleted ammonia. Fish therefore should enrich sediment ^{15}N values, which is the opposite of what we observed.

There is some evidence that algae, particularly diatoms, fractionate N upon uptake of NO_3 and NH_4. Isotopic fractionation of NO_3 in field studies and laboratory cultures have reported depletion of $\delta^{15}N$ in marine diatoms by −4 to −5‰, to as low as −24‰ (Pennock and others 1996; Wada and Hattori 1978). Additional, comparative studies of fractionation in diatoms and other types of algae conclude that the isotopic disturbance associated with phytoplankton blooms is greatest when diatoms are the dominant species (Montoya and McCarthy 1995). The coincident depletion of sediment $\delta^{15}N$ with the dominance and abundance of *A. formosa* could very well reflect, then, eutrophication and physiological changes in N processing due to the shift in species composition. A tributary stream to the Loch shows a range of $\delta^{15}N$ from −2 to 5‰ (Kendall and others 1995), but our information is not complete enough to calculate a hypothetical fractionation rate from algae to compare with sediment values.

Approximately half of the wet N deposition to high Front Range locations is NH_4, almost certainly coming from an agricultural source. Nitrogen oxides also can have agricultural sources, and most fertilizers applied to US agricultural fields today are commercially produced from atmospheric N through the Haber-Bosch process. Fertilizer N therefore has an initial signature of 0 ± 3‰ (Smil 1997; Kendall 1998). Ammonia gas additionally volatilizes from animal waste, and 50–80% of animal waste from commercial feedlots spread on fields volatilizes within the first 48 hours (Lockyer and others 1989; Sharpe and Harper 1997). Volatilized NH_3 is isotopically light, with values that range to −17‰ (Kerley and Jarvis 1996; D. Rowan, personal communication). Colorado ranks fourth in the United States for confined feeding operations and is tenth in the nation in beef production, much of which occurs in the South Platte Basin (Colorado Agricultural Statistics Service 1999). An influx of isotopically light N in wet deposition therefore is a logical explanation for our observed sediment N signatures.

Similarities to other studies. Although our examination of sediment N isotopic signatures is the first to our knowledge, there are two plausible explanations for depletion of $\delta^{15}N$ since approximately

1950: a shift in algal communities to one now dominated by the diatom *A. formosa*, and introduction of isotopically lighter N in atmospheric deposition due to a changing mixture of N sources. The diatom fractionation work cited above supports the hypothesis that a shift in communities could cause the observed shift in sediment isotopic signature. We know of no historical studies of $\delta^{15}N$ in deposition, but an isotopic characterization of rain and North Carolina estuaries and coastal waters showed the waters became more negative after prolonged periods of rain, corresponding with abundant inputs of isotopically light NO_3 (average of +1.0‰), and NH_4 (average of −3.1‰; Paerl and Fogel 1994). The coastal plain of North Carolina leads the United States in hog production, mostly in confined feeding operations. Both deposition and algal community changes are indicative of human-induced changes, because alterations of algal communities were initiated by introduction of nutrients (N, P, or both).

Time Frame for Ecological Change

Rates of change in both sediment diatom communities and isotopic signatures are greatest since 1950. Furthermore, at least 75% of the difference in amount of N stored in the soil organic matter pool between east and west side stands can be accounted for if N deposition rates increased with population growth. Although there is a trend of increasing N in precipitation, the contribution of this increase over the past 10 years comes to 0.26 kg N ha^{-1} y^{-1}. Lake N concentrations since 1983 show no evidence of trends. Taken together, these data suggest the greatest chemical change to alpine and subalpine ecosystems occurred before our intensive monitoring.

The post-1950 period of rapid sediment isotopic shifts, diatom community and productivity changes, and soil N accumulation corresponds with intensification of agricultural practices, animal husbandry, and population growth in the South Platte River Basin east of the Front Range (Figure 5). Cattle numbers in confined feedlots rose from 1.4 to 3.7 million head between 1940 and 1973, whereas human population in the 10 counties of the South Platte increased from 713,000 to 2.4 million between 1950 and 1995 (Colorado Agricultural Statistics Service 1990; US Census Bureau 1998). Increasing availability and application of commercial fertilizers began around 1950, leading to massive introductions of synthetic N to cultivated soils (Alexander and Smith 1990). Automobile transport, energy consumption, animal waste, and profligate fertilizer use all contribute N emissions to the atmosphere for transport to the mountains. Other investigators also have noted the increase in fixed and agricultural N emissions to the atmosphere beginning around 1950, so the trends from our study area seem to be indicative of a larger number of regions worldwide where agricultural and urban activities expanded during the past 50 years (Mayewski and others 1986, 1990; Arbaugh et al. 1999).

Conclusions

Taken individually, our observations of forest and soil properties, recent shifts in abundance and composition of diatom communities, and trends in N isotopic signatures have plausible explanations other than N deposition. Diatom community shifts could be due to episodic P introductions; the isotopic N record in sediments could reflect preferential assimilation of light N by *A. formosa*. But these explanations do not hold up well when compared with the rest of our observations: east side N deposition is elevated and increasing relative to west side deposition; east side lakes are enriched in NO_3 over west side lakes. Forest and soil biogeochemical characteristics are in line with other reports, albeit the east side Colorado stands are at the low end of the response spectrum relative to many of these studies.

Several of our observations suggest the east side of the Front Range is at the beginning of a trajectory of change. The absence of changes in soil potential net nitrification, foliar Ca:Al, and foliar lignin suggest changes have only begun to occur. The abundance of *A. formosa* and *F. crotonensis* in recent lake sediments reflects the onset of human-caused disturbance. Our results strongly suggest Rocky Mountain ecosystems are extremely sensitive and responsive to slight increases in N availability.

Acknowledgments

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References

Aber J, McDowell W, Nadelhoffer K, Magill A, Berntson G, Kamakea M, McNulty S, Currie W, Rustad L, Fernandez I (1998) Nitrogen saturation in temperate forest ecosystems: hypothesis revisited. Bioscience 48:921–34

Alexander RB, Smith RA (1990) County level estimates of nitrogen and phosphorus fertilizer use in the United States, 1945–1985. Reston, VA: US Geological Survey Open-file Report nr 90–130

Alexander RR, Watkins RK (1977) The Fraser Experimental Forest, Colorado. USDA Forest Service General Technical Report nr RM-40. Fort Collins, CO. 32 p

Anderson NJ (1994) Comparative planktonic diatom biomass responses to lake and catchment disturbance. J Plankton Res 16:133–50

Anderson NJ, Renberg I, Segerstrom U (1995) Diatom production responses to the development of early agriculture in a boreal forest catchment (Kassjön, northern Sweden). J Ecol 83:809–22

Appleby PG, Oldfield F (1978) The calculation of lead-210 dates assuming a constant rate of supply of unsupported 210Pb to the sediment. Catena 5:1–18

Arbaugh MJ, Peterson DL, Miller PR (1999) Air pollution effects on growth of ponderosa pine, Jeffrey pine, and bigcone Douglas fir. In: Miller PR, McBride JR, editors. Oxidant air pollution impacts in the montane forests of Southern California. New York: Springer-Verlag. p 179–207

Axler RP, Rose C, Tikkanen CA (1994) Phytoplankton nutrient deficiency as related to atmospheric nitrogen deposition in northern Minnesota acid-sensitive lakes. Can J Fish Aquat Sci 51:1281–96

Baker LA, Kaufmann PR, Herlihy AT, Eilers JM (1990) Current status of surface water acid-base chemistry. State of Science/Technology, Report nr 9. Washington, DC: National Acid Precipitation Assessment Program

Baron JS (1983) Comparative water chemistry of four lakes in Rocky Mountain National Park. Water Resources Bull 19:897–902

Baron JS, editor (1992) Biogeochemistry of a subalpine ecosystem: Loch Vale Watershed. Ecological Study Series #90. New York: Springer-Verlag

Baron JS, Caine N (2000) Temporal coherence of two alpine lake basins of the Colorado Front Range, U.S.A. Freshwater Biology. 43:463–476

Baron JS, Campbell DH (1997) Nitrogen fluxes in a high elevation Colorado Rocky Mountain basin. Hydrol Processes 11: 783–99

Baron JS, Denning AS (1993) The influence of mountain meteorology on precipitation chemistry at low and high elevations of the Colorado Front Range, U.S.A. Atmospheric Environ 27A:2337–49

Baron J, Ojima DS, Holland EA, Parton WJ (1994) Nitrogen consumption in high elevation Rocky Mountain tundra and forest and implications for aquatic systems. Biogeochemistry 27:61–82

Baron JS, Hartman MD, Band LE, Lammers RB (2000) Sensitivity of a high elevation Rocky Mountain watershed to altered climate and CO_2. Water Resources Res 36:89–99

Battarbee RW (1973) A new method for estimation of absolute microfossil numbers, with reference especially to diatoms. Limnol Oceanogr 18:647–53

Battarbee RW (1986) Diatom analysis. In: Berglund BE, editor. Handbook of holocene palaeoecology and palaeohydrology. Chichester, UK: Wiley Interscience. p 527–70

Bossert JE (1990) Regional-scale flows in complex terrain: an observational and numerical investigation [dissertation]. Fort Collins, CO: Department of Atmospheric Science, Colorado State University. 257 p

Bowman WD, Theodore TA, Schardt JC, Conant RT (1993) Constraints of nutrient ability on primary production in two alpine tundra communities. Ecology 74:2085–97

Brabrand A, Faafeng BA, Nilssen JPM (1990) Relative importance of phosphorus supply to phytoplankton production: fish excretion versus external loading. Can J Fish Aquat Sci 47:364–72

Campbell DH, Clow DW, Ingersoll GP, Mast MA, Spahr NE, Turk JT (1995) Temporal variations in the chemistry of 2 snowmelt-dominated streams in the Rocky Mountains. Water Resources Res 31:2811–22

Carpenter SR, Kitchell JF, Hodgson JR (1985) Cascading trophic interactions and lake productivity. BioScience 35:634–9

CASTnet (1999) Clean Air Status and Trends Network. Washington, DC: United State Environmental Protection Agency. http://www.epa.gov/acidrain/CASTNET/

Christie CE, Smol JP (1993) Diatom assemblages as indicators of lake trophic status in southeastern Ontario lakes. J Phycol 29:575–86

Clow DW, Mast MA (1999) Long-term trends in stream water and precipitation chemistry at five headwater basins in the northeastern United States. Water Resources Res 35:541–54

Cole JJ, Caraco NF, Likens GE (1990) Short-range atmospheric transport: a significant source of phosphorus to an oligotrophic lake. Limnol Oceanogr 35:1230–8

Colorado Agricultural Statistics Service Annual Bulletins since 1925. Issued cooperatively by the National Agricultural Statistics Service and the Colorado Department of Agriculture. Published by Colorado Agricultural Statistics Service, Denver CO. Denver: Colorado Department of Agriculture, Colorado Agricultural Statistics Service, Denver, CO

Daly C, Neilson RP, Phillips DL (1994) A statistical-topographic model for mapping climatological precipitation over mountainous terrain. J Appl Meteorol 33:140–58

Denning AS, Baron J, Mast MA, Arthur MA (1991) Hydrologic pathways and chemical composition of runoff during snowmelt in Loch Vale Watershed, Rocky Mountain National Park, Colorado USA. Water Air Soil Pollut 59:107–23

Dise NB, Wright RF (1995) Nitrogen leaching from European forests in relation to nitrogen deposition. Forest Ecol Manage 71:153–61

Eilers JM, Kanciruk P, McCord RA, Overton WS, Hook L, Blick DJ, Brakke DF, Kellar PE, DeHaan MS, Silverstein ME, and others (1987) Characteristics of lakes in the western United States. Volume II. Data compendium for selected physical and chemical variables. EPA 600/3-86/054b. Washington, DC: US Environmental Protection Agency

Elser JJ, Marzolf ER, Goldman CR (1990) Phosphorus and nitrogen limitation of phytoplankton growth in the freshwaters of North America: a review and critique of experimental enrichments. Limnol Oceanogr 24:401–16

Emmett BA, Boxman D, Bredemeier M, Gundersen P, Kjønass OJ, Moldan F, Schleppi P, Teitema A, Wight RF (1998) Predicting the effects of atmospheric nitrogen deposition on conifer stands: evidence from the NITREX ecosystem-scale experiments. Ecosystems 1:352–60

[EPA] Environmental Protection Agency (1987) Handbook for methods for acid deposition studies. Laboratory analyses for surface water chemistry. EPA 600/4-87/026. Washington, DC: US Environmental Protection Agency

Fenn ME, Poth M, Aber JD, Baron JS, Bormann BT, Johnson

DW, Lemly AD, McNulty SG, Ryan DF, Stottlemyer R (1998) Nitrogen excess in North American ecosystems: a review of predisposing factors, geographic extent, ecosystem responses, and management strategies. Ecol Appl 8:706–33

Foged N (1981) Diatoms in Alaska. Vaduz, Germany: J. Cramer Verlag

Friedland AJ, Hawley GJ, Gregory RA (1988) Red spruce (*Picea rubens* Sarg.) foliar chemistry in northern Vermont and New York, USA. Plant Soil 105:189–93

Fry B, Brand W, Mersch FJ, Tholke K, Garritt R (1992) Automated analysis system for coupled $\delta^{13}C$ and $\delta^{15}N$ measurements. Analyt Chem 64:288–91

Germain H (1981) Flore des diatomées, eaux douces et saumâtres du Massif Armericain et des contrées voisines d'Europe occidentale. Paris: Société Nouvelle des Éditions Boubée

Glew JR (1988) A portable extruding device for close interval sectioning of unconsolidated core samples. J Paleolimnol 1:235–9

Glew JR (1989) A new trigger mechanism for sediment samplers. J Paleolimnol 2:241–3

Gundersen P, Rasmussen L (1990) Nitrification in forest soil: effects of nitrogen deposition on soil acidification and aluminum release. Rev Environ Contaminants Toxicol 113:1–45

Gundersen P, Emmett BA, Kjonaas OJ, Koopmans CJ, Tietema A (1998) Impact of nitrogen deposition on nitrogen cycling in forests: a synthesis of NITREX data. For Ecol Manage 101:37–56

Hall RI, Leavitt PR, Quinlan R, Dixit AS, Smol JP (1999) Effects of agriculture, urbanization, and climate on water quality in the northern Great Plains. Limnol Oceaongr 44:739–56

Hastie TJ, Tibshirani RJ (1990) Generalized additive models. London: Chapman and Hall

Hunt HW, Ingham ER, Coleman DC, Elliott ET, Reid CPP (1988) Nitrogen limitation of production and decomposition in prairie, mountain meadow and pine forest. Ecology 69:1009–16

Jacobson GL Jr, Grimm EC (1986) A numerical analysis of Holocene forest and prairie vegetation in central Minnesota. Ecology 67:958–66

Jassby AD, Reuter JE, Axler RP, Goldman CR, Hackley SH (1994) Atmospheric deposition of nitrogen and phosphorus in the annual nutrient load of Lake Tahoe (California-Nevada). Water Resources Res 30:2207–16

Johnson DW, Lindberg SE, editors (1992) Atmospheric deposition and forest nutrient cycling. Ecological studies 91. New York: Springer-Verlag

Kahl JS, Norton SA, Fernandez IJ, Nadelhoffer KJ, Driscoll CT, Aber JD (1993) Experimental inducement of nitrogen saturation at the watershed scale. Environ Sci Technol 27:565–8

Kendall C (1998) Tracing nitrogen sources and cycles in catchments. In: Kendall C, McDonnell JJ, editors. Isotope tracers in catchment hydrology. New York: Elsevier. p 519–76

Kendall C, Campbell DH, Burns DA, Shanley JB, Silva SR, Chang CCY (1995) Tracing sources of nitrate in snowmelt runoff using the oxygen and nitrogen isotopic compositions of nitrate. In: Biogeochemistry of Seasonally Snow-Covered Catchments. Tonnessen KA, Williams MW, Tranter M, editors. IAHS Publication nr 228. Wallingford, Oxfordshire, UK: IAHS Press. p 339–47

Kerley SJ, Jarvis SC (1996) Preliminary studies of the impact of excreted N on cycling and uptake of N on pasture systems using natural abundance isotope discrimination. Plant Soil 178:287–94

Krammer K, Lange-Bertalot H (1986–1991) Bacillariophyceae 1 Teil: Naviculaceae (1986); 2 Teil: Bacillariaceae, Epithemiaceae, Surirellaceae, (1988); 3 Teil: Centrales, Fragilariaceae, Eunotiaceae, (1991a); 4 Teil: Achnanthaceae, Kritische Ergäänzungen zu Navicula (Lineolatae) und Gomphonema Gesamtliteraturverzeichnis, (1991b). In: Ettl H, Gerloff H, Heynig H, Mollenhauer D, editors. Sübwasserflora von Mitteleuropa 2/1–4. Stuttgart: Gustav Fischer Verlag

Langford AO, Fehsenfeld FC (1992) Natural vegetation as a source or sink for atmospheric ammonia: a case study. Science 255:581–3

Leavitt PR, Schindler DE, Paul AJ, Hardie AK, Schindler DW (1994) Fossil pigment records of phytoplankton in trout-stocked alpine lakes. Can J Fish Aquat Sci 51:2411–23

Lewis WM Jr (1982) Changes in pH and buffering capacity of lakes in the Colorado Rockies. Limnol Oceanogr 27:167–72

Lewis WM Jr, Grant MC (1979) Changes in the output of ions from a watershed as a result of the acidification. Ecology 60:1093–7

Lewis WM Jr, Grant MC (1980) Acid precipitation in the western U.S. Science 207:176–7

Lewis, WM Jr., Grant MC, Saunders JF, III (1984) Chemical patters of bulk atmospheric deposition in the state of Colorado. Water Resources Res 20:1691–1704

Likens GE, Driscoll CT, Buso DC (1996) Long-term effects of acid rain: response and recovery of a forest ecosystem. Science 272:244–6

Lockyer DR, Pain BF, Klarenbeek JV (1989) Ammonia emissions from cattle, pig, and poultry wastes applied to pasture. Environ Pollut 56:19–30

Lotter A (1998) The recent eutrophication of Balderggersee (Switzerland) as assessed by fossil diatom assemblages. Holocene 8:395–405

Lovering TS, Goddard EN (1959) Geology and ore deposits of the Front Range, Colorado. U.S. Geological Survey Professional Paper nr 223. Denver, CO: USGS

Lynch JA, Grimm JW, Bowersox VC (1995) Trends in precipitation chemistry in the United States: a national perspective, 1980–1992. Atmospheric Environment 29:1231–1246

Magill AH, Aber JD, Hendricks JJ, Bowden RD, Melillo JM, Steudler PA (1997) Biogeochemical response of forest ecosystems to simulated chronic nitrogen deposition. Ecol Appl 7:402–15

Mayewski PA, Lyons WB, Spencer MJ, Twickler M, Dansgaard W, Koci B, Davidson CI, Honrath RE (1986) Sulfate and nitrate concentrations from a south Greenland ice core. Science 232:975–7

Mayewski PA, Lyons WB, Spencer MJ, Twickler MS, Buck CF, Whitlow S (1990) An ice-core record of atmospheric response to anthropogenic sulfate and nitrate. Science 346:554–6

McDonnell MJ, Pickett STA, editors (1993) Humans as components of ecosystems: the ecology of subtle human effects and populated areas. New York: Springer-Verlag

McKnight DM, Smith RL, Bradbury JP, Baron JS, Spaulding S (1990) Phytoplankton dynamics in three Rocky Mountain lakes, Colorado, USA. Arctic Alpine Res 22:264–74

McNulty SG, Aber JD, Boone RD (1991) Spatial changes in forest floor and foliar chemistry of spruce-fir forests across new England. Biogeochemistry 14:13–29

Menounos B, Reasoner MA (1997) Evidence for cirque glaciation in the Colorado Front Range during the Younger Dryas chronosequence. Quaternary Res 48:38–47

Montoya JP, McCarthy JJ (1995) Isotpic fractionation during nitrate uptake by phytoplankton grown in continuous-culture. J Plankton Res 17:439–64

Morris DP, Lewis WM Jr (1988) Phytoplankton nutrient limitation in Colorado mountain lakes. Freshwater Biol 20:315–27

Musselman RC, Hudnell L, Williams MW, Sommerfeld RA (1996) Water chemistry of Rocky Mountain front Range aquatic ecosystems. Research Paper nr RM-RP-325. Fort Collins, CO: US Department of Agriculture, Forest Service, Rocky Mountain Forest and Range Experiment Station. 13 pp

[NADP]/NTN Database National Atmospheric Deposition Program (NRSP-3)/National Trends Network (1999) Champaign, IL: NADP Program Office, Illinois State Water Survey

Paerl HW, Fogel ML (1994) Isotopic characterization of atmospheric nitrogen inputs as sources of enhanced primary production in coastal Atlantic-ocean waters. Mar Biol 119:635–45

Parrish DD, Trainer M, Williams EJ, Fahey DW, Hubler G, Eubank SC, Liu SC, Murphy PC, Albritton DL, Fehsenfeld FC (1990) Measurements of the Nox-O_3 photostationary state at Niwot Ridge, Colorado. J Geophys Res 91:5361–70

Patrick R, Reimer CW (1966) The diatoms of the United States Exclusive of Alaska and Hawaii. Monograph 13, volume 1. published by Academy of Natural Sciences of Philadelphia, Philadelphia, PA. 688 p

Patrick R, Reimer CW (1975) The diatoms of the United States Exclusive of Alaska and Hawaii. Monograph 13, Volume 1. published by Academy of Natural Sciences of Philadelphia, Philadelphia, PA. 578 p

Peet RK (1988) Forests of the Rocky Mountains. In: Barbour MG, Billings WD, editors. North American terrestrial vegetation. Cambridge, MA: Cambridge University Press. p 63–102

Pennock JR, Velinsky DJ, Ludlan JM, Sharp JH, Fogel ML (1996) Isotopic fractionation of ammonium and nitrate during uptake of *Skeletonema costatum*: implications for $\delta^{15}N$ dynamics under bloom conditions. Limnol Oceanogr 41:451–9

Prescott CE (1995) Does nitrogen availability control rates of litter decomposition in forests? Plant Soil 168–169:83–8

Renberg I, Korsman T, Anderson NJ (1993) A temporal perspective of lake acidification in Sweden. Ambio 22:264–71

Reuss JO, Johnson DW (1986) Acid deposition and the acidification of soil and waters. Ecological Studies 59. New York: Springer-Verlag

Rosenlund BD, Stevens DR (1990) Fisheries and aquatic management: Rocky Mountain National Park 1988–1989. Golden, CO: US Fish and Wildlife Service, Colorado Fish and Wildlife Assistance Office

Schaefer DA, Driscoll CT, van Dreason R, Yatsko CP (1990) The episodic acidification of Adirondack lakes during snwomelt. Water Resources Res 26:1639–47

Schlesinger WH (1997) Biogeochemistry: ana analysis of global change (2nd ed. New York: Academic Press

Schulze E-D, Lange OL, Oren R (1989) Forest decline and air pollution. Berlin: Springer-Verlag

Sharpe RR, Harper LA (1997) Ammonia and nitrous oxide emissions from sprinkler irrigation applications of swine effluent. J Environ Qual 26:1703–6

Sievering H, Rusch D, Marquez L (1996) Nitric acid, particulate nitrate and ammonium in the continental free troposphere: nitrogen deposition to an alpine tundra ecosystem. Atmospheric Environ 30:2527–37

Smil V (1997) Global population and the nitrogen cycle. Sci Am 277:76–81

Soderstrom B, Baath E, Lundgren B (1983) Decrease in soil microbial activity and biomasses owing to nitrogen amendments. Revue Can Microbiol 29:1500–6

Stoddard JL (1994) Long-term changes in watershed retention of nitrogen: its causes and quatic consequences. In: Baker LA, editor. Environmental chemistry of lakes and reservoirs. Advances in Chemistry Series No. 237. Washington, DC: American Chemical Society. p223–284

Stoddard JL, Jeffries DS, Lükewille A, Clair TA, Dillon PJ, Driscoll CT, Forsius M, Johannessen M, Kahl JS, Kellogg JH and others (1999) Regional trends in aquatic recovery from acidification in North America and Europe. Nature 401:575–8

Stottlemyer R, Troendle CA, Markowitz D (1997) Change in snowpack, soil water, and streamwater chemistry with elevation during 1990, Fraser Experimental Forest, Colorado. J Hydrol 195:114–36

Tietema A (1998) Microbial carbon and nitrogen dynamics in coniferous fourest floor material collected along a European nitrogen deposition gradient. For Ecol Manage 101:29–36

Tietema A, Beier C (1995) A correlative evaluation of nitrogen cycling in the forest ecosystems of the EC projects NITREX and EXMAN. For Ecol Manage 71:143–51

US Census Bureau (1997) Population distribution and population estimates branches, United States Department of Commerce, http://www.census.gov/population/www/estimates/popest.html/

Veblen TT, Lorenz DC (1991) The Colorado Front Range: a century of ecological change. Salt Lake City (UT) University of Utah Press. 186 pp

Vitousek PM, Aber JD, Howarth RW, Likens GE, Matson PA, Schindler DW, Schelsinger WH, Tilman DG (1997) Human alteration of the global nitrogen cycle: sources and consequences. Ecol Appl 7:737–50

Wada E, Hattori A (1978) Nitrogen isotope effects in the assimilation of inorganic nitrogenous compounds by marine diatoms. Geomicrobiology 1:85–101

Williams MW, Baron JS, Caine N, Sommerfeld R, Sanford R (1996a) Nitrogen saturation in the Rocky Mountains. Environ Sci Technol 30:640–6

Williams MW, Losleben M, Caine N, Greenland D (1996b) Changes in climate and hydrochemcial responses in a high-elevation catchment in the Rocky Mountains, USA. Limnol Oceanogr 41:939–46

Williams MW, Bardsley T, Rikkers M (1998) Overestimation of snow depth and inorganic nitrogen wetfall using NADP data, Niwot Ridge, Colorado. Atmospheric Environ 32:3827–33

Wolfe AP (1997) On diatom concentrations in lake sediments: results of an interlaboratory comparison and other experiments performed on a uniform sample. J Paleolimnol 18:61–6