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Atmospheric nitrogen deposition in the Rocky Mountains of Colorado and southern Wyoming—a review and new analysis of past study results

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Abstract

The Rocky Mountain region of Colorado and southern Wyoming receives as much as 7 kg ha^{-1} yr^{-1} of atmospheric nitrogen (N) deposition, an amount that may have caused changes in aquatic and terrestrial life in otherwise pristine ecosystems. Results from published studies indicate a long-term increase in the rate of atmospheric N deposition during the 20th century, but data from the National Atmospheric Deposition Program and Clean Air Status and Trends Network show no region-wide increase during the past 2 decades. Nitrogen loads in atmospheric wet deposition have increased since the mid-1980s, however, at three high elevation (>3000 m) sites east of the Continental Divide in the Front Range. Much of this increase is the result of increased ammonium (NH_4^+) concentrations in wet deposition. This suggests an increase in contributions from agricultural areas or from vehicles east of the Rocky Mountains and is consistent with the results of previous studies that have suggested a significant eastern source for atmospheric N deposition to the Front Range. The four sites with the highest NH_4^+ concentrations in wet deposition were among the six easternmost NADP sites, which is also consistent with a source to the east of the Rockies. This analysis found an increase in N loads in wet deposition at Niwot Ridge of only 0.013 kg ha^{-1} yr^{-1}, more than an order of magnitude less than previously reported for this site. This lower rate of increase results from application of the non-parametric Seasonal Kendall trend test to mean monthly data, which failed a test for normality, in contrast to linear regression, which was applied to mean annual data in a previous study. Current upward trends in population growth and energy use in Colorado and throughout the west suggest a need for continued monitoring of atmospheric deposition of N, and may reveal more widespread trends in N deposition in the future.

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Keywords: Atmospheric deposition; Nitrogen; Colorado; Rocky mountains; Nitrogen saturation

1. Introduction

Human activities modify and accelerate the global cycle of nitrogen (N). Fixation of N by humans for energy production, fertilizer production, and crop cultivation now exceeds the amount of biologically fixed N on the continents (Galloway et al., 1995). One aspect of human alteration of the N cycle is the release of NO_x gas from fossil fuel combustion and NH_3 gas from agricultural production to the atmosphere, where they may then be converted to nitrate (NO_3^-) and ammonium (NH_4^+) respectively, and deposited on the land surface as wet and dry deposition (Vitousek et al., 1997). Particulate forms of N are formed from precursor emissions by human activities and are also emitted and transported through the atmosphere to be deposited later as dry deposition. The rate of atmospheric N deposition has been greatly increased through human

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activities such as burning fossil fuels and fertilizer use, and high rates of atmospheric N deposition have been widely documented in Europe as well as in North America (Lovett, 1994; Fenn et al., 1998; Lawrence et al., 2000).

The Rocky Mountain region of Colorado and southern Wyoming receives 2–4 kg ha^{-1} yr^{-1} of atmospheric N in wet deposition (National Atmospheric Deposition Program [NADP], 2000), and rates as high as 5.5 kg ha^{-1} yr^{-1} have been reported for the Loch Vale watershed in Rocky Mountain National Park (Fig. 1) (Campbell et al., 2000). These rates are lower than those reported for the midwestern, southeastern, and northeastern parts of the United States (NADP, 2000), however, symptoms of advanced stages of N saturation have been reported in alpine ecosystems of the Front Range of the Rocky Mountains (Baron et al., 1994; Williams et al., 1996a; Williams and Tonnessen, 2000). The thin and sparse soil and the lack of forest vegetation at high elevations in alpine watersheds of the Colorado Rockies result in the export of a large proportion of the N in atmospheric deposition, and the apparent sensitivity of alpine ecosystems to atmospheric N deposition. Retention rates of atmospheric N deposition in subbasins of the Loch Vale watershed range from only 19% to 60% (Campbell et al., 2000). Wet deposition of NO$_3^-$ increased from the mid-1980s to the mid-1990s at the Niwot Ridge NADP site (Fig. 1), and a similar increase in stream NO$_3^-$ concentrations during the growing season was reported for the outlet of Green Lake 4, near Niwot Ridge (Fig. 1) (Williams et al., 1996a). The increased deposition of atmospheric N in the Front Range may adversely affect amphibian populations, and alter terrestrial plant community composition, foliar

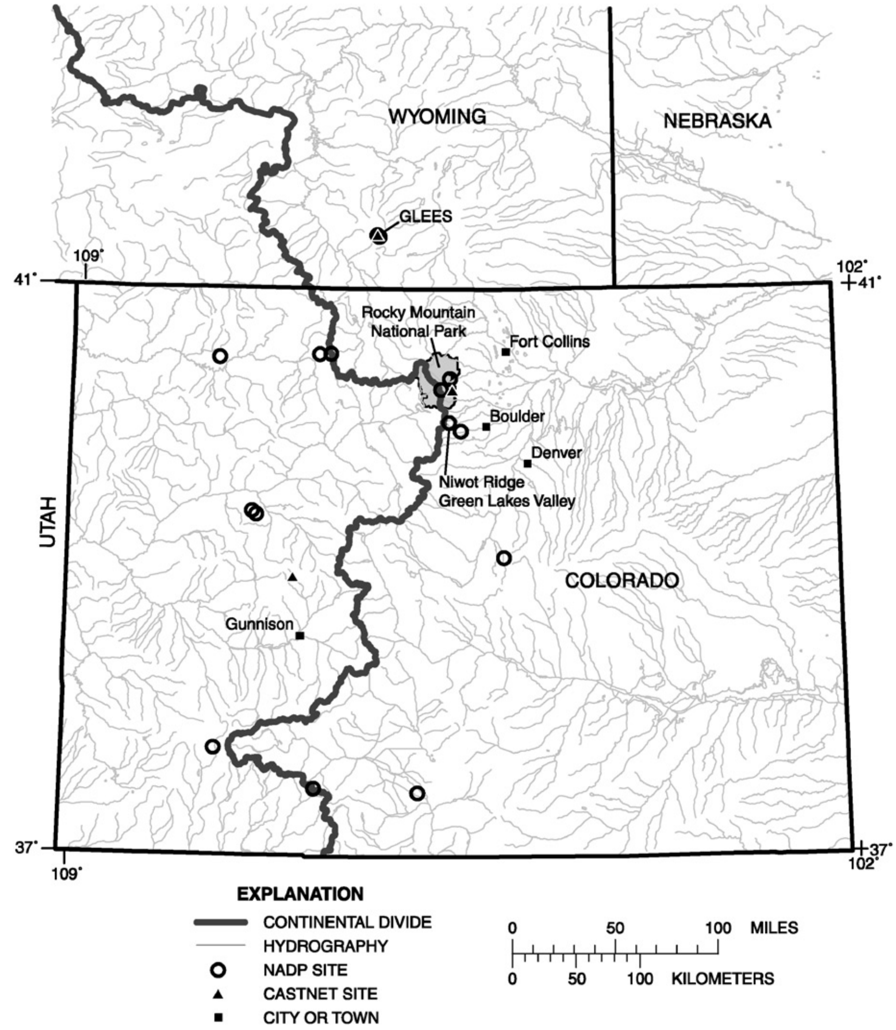

Fig. 1. Map showing field sites discussed in report, including cities, NADP and CASTNET sites, streams and lakes, sites of intensive field studies, and the Continental Divide.

N:P in bristlecone pines, soil-bacteria and fungal communities, and phytoplankton dynamics (Mancinelli, 1986; Morris and Lewis, 1988; Harte and Hoffman, 1989; Bowman and Steltzer, 1998; Williams et al., 1996a).

An increased interest in the effects of atmospheric N deposition in the Colorado and Wyoming Rockies is evident among researchers based on the large number of publications on the subject during the past decade (Baron, 1992; Baron et al., 1994; Baron and Campbell, 1997; Brooks and Williams, 1999; Brooks et al., 1999; Campbell et al., 2000; Meixner et al., 2000, many others). In addition to interest in the effects of atmospheric N deposition in the Colorado Rockies among researchers, there is also significant interest among Federal land managers because the region includes the Rocky Mountain National Park and several other wilderness areas and wildlife refuges that are protected by law from damage by air pollution under provisions of the Clean Air Act Amendments of 1977. These Federal land managers are responsible for protecting air-quality related values in these Class 1 wilderness areas.

Given the interest and the potential ecological significance of atmospheric N deposition in the Colorado Rockies, researchers need an accurate picture of the rates of deposition, geographic and topographic sources of variation in those rates, and whether temporal trends are present or not. The purpose of this paper is to review existing literature on atmospheric N deposition in the Rocky Mountains of Colorado and southern Wyoming, analyze current patterns and trends in N deposition, and to perform a critical assessment of past studies based on this new analysis.

The principal geographic focus of this review and new analysis is the Colorado Rocky Mountains and intermontane area, and the area surrounding the Glacier Lakes Ecosystem Experiments site (GLEES) in adjacent southeastern Wyoming (Fig. 1). The Wyoming study area is included here because atmospheric deposition patterns appear to be similar to those of nearby Colorado, there is a history of atmospheric deposition data collection that goes back to the 1980s, and because a wealth of publications are available based on data collected in the area (Musselman, 1994).

2. Methods

Relevant publications were reviewed and are cited in appropriate sections of the paper. Additionally, original data on wet and dry deposition of N were retrieved and analyzed. Time trends in rates and concentrations of atmospheric wet and dry deposition of N were analyzed through the Seasonal Kendall non-parametric statistical test (Helsel and Hirsch, 1992). These atmospheric-deposition data were collected as part of the National Atmospheric Deposition Program (NADP) and the Clean Air Status and Trends Network (CASTNET). The NADP uses weekly samples of precipitation from wet-dry collectors that are sent to a laboratory at the Illinois State Water Survey for chemical analysis. Details about methods and QA procedures are available at http://nadp.sws.uiuc.edu/QA. The CASTNET uses weekly mean concentrations of NO_3^-, NH_4^+, and HNO_3 from filter packs that are retrieved from towers. Each CASTNET site also collects meteorological data and uses information on local vegetation and land use to calculate dry deposition of N according to a “big leaf” model (Baldocchi, 1988). Filter packs are extracted and analyzed at Harding ESE laboratory in Gainesville, FL. Details of the assumptions and methods used to estimate dry deposition are available at http://www.epa.gov/castnet and in Clarke et al. (1997).

3. Results and discussion

Data are summarized on current rates of atmospheric deposition of NO_3^- and NH_4^+, and concentrations of these ions in precipitation from 15 sites in the region that are part of NADP and 3 sites that are part of CASTNET (Fig. 1). Additionally, time trends in concentrations and loads are examined. These data are compared to the results of previous studies of atmospheric N deposition in the region. Finally, the source areas of the N in atmospheric deposition (east or west of the Rockies) are examined as reported in previously published studies.

3.1. Concentrations and loads of inorganic nitrogen in wet deposition

Precipitation samples are collected weekly at 15 NADP sites in the Rocky Mountain region of Colorado and southern Wyoming (Fig. 1) (data available at http://nadp.sws.uiuc.edu). The length of data collection at these sites varies from 8 to 21 yr, and 10 sites have a data record of between 12 and 17 yr. During 1998, the chemistry of bulk deposition in and around Rocky Mountain National Park was monitored at 15 additional sites (Ingersoll et al., 2000). Additionally, the snowpack was sampled regularly at 27 sites in Colorado and southern Wyoming in March–April of 1993–97 (Turk et al., 2001).

The mean annual load (1995–99) of inorganic N in wet deposition at the 15 NADP sites ranges from 0.7 kg N ha^{-1} yr^{-1} at the Alamosa site to 6.8 kg N ha^{-1} yr^{-1} at the Niwot Saddle site (Table 1). The precipitation collector at Niwot Saddle catches blowing snow in excess of the amount deposited, however, necessitating a 32% downward correction in annual inorganic N loads for 1986–95 (Williams et al.,

Table 1
Mean annual wet deposition of N, precipitation amount, NO_3^- concentration, and NH_4^+ concentrations for 1995–99 at 15 National Atmospheric Deposition Program (NADP) sites in the Rocky Mountains of Colorado and southern Wyoming

Site name	Elev. (m)	N deposition ($kg\,N\,ha^{-1}\,yr^{-1}$)	Precipitation (cm)	NO_3^- ($\mu mol\,l^{-1}$)	NH_4^+ ($\mu mol\,l^{-1}$)
East of Divide					
Alamosa	2298	0.7 (0.06)	16.8 (2.1)	13.7 (1.5)	16.9 (4.3)
Beaver Meadows	2490	2.0 (0.3)	47.4 (7.3)	16.9 (1.3)	14.0 (1.9)
Brooklyn Lake	3212	2.7 (0.3)	131.2 (10.7)	9.7 (1.1)	4.7 (0.5)
Loch Vale	3159	3.0 (0.3)	120.7 (21.4)	11.7 (2.4)	6.7 (0.8)
Manitou	2362	2.1 (0.1)	45.4 (7.1)	21.1 (3.0)	12.5 (2.4)
Niwot Saddle	3520	4.6[a] (1.3)	157.8 (42.0)	14.0 (0.2)	7.0 (1.3)
Snowy Range	3286	3.7 (0.5)	134.4 (22.4)	12.2 (1.4)	6.8 (1.1)
Sugarloaf	2524	2.8 (0.5)	57.8 (8.3)	18.3 (2.4)	16.0 (3.5)
At Divide					
Buffalo Pass	3234	3.3 (0.5)	134.4 (22.4)	11.4 (1.4)	6.1 (1.0)
Wolf Creek Pass	3292	3.3 (0.5)	143.7 (10.0)	11.4 (1.0)	5.0 (1.0)
West of Divide					
Dry Lake	2527	2.5 (0.4)	93.3 (14.2)	13.2 (1.5)	6.1 (1.2)
Four Mile Park	2502	1.5 (0.3)	61.5 (13.3)	11.1 (1.9)	6.3 (1.0)
Molas Pass	3249	1.8 (0.2)	85.5 (5.9)	10.9 (2.4)	4.2 (0.8)
Sand Spring	1998	1.2 (0.2)	36.3 (6.7)	15.3 (2.7)	7.8 (1.5)
Sunlight Peak	3206	1.6 (0.2)	73.6 (8..7)	10.6 (1.7)	5.6 (0.8)

[a] Corrected by subtracting 32% of the measured total according to Williams et al. (1998).
Standard deviation of the mean is in parentheses.

1998). If the 1995–99 mean for this site is adjusted similarly, the resulting mean load of $4.6\,kg\,N\,ha^{-1}\,yr^{-1}$ is still the highest in the region, and close to the values of 3.0–3.7 calculated for the Buffalo Pass, Loch Vale, Snowy Range, and Wolf Creek Pass sites (Table 1). All NADP sites with mean annual wet deposition loads of less than $2.0\,kg\,N\,ha^{-1}\,yr^{-1}$ are below elevations of 3000 m or are west of the Continental Divide. The mean annual inorganic N load in wet deposition at the 15 NADP sites is highly correlated with precipitation amount ($r^2 = 0.59$, $p = 0.003$, as indicated by least-squares linear regression) but is not correlated with the mean annual dissolved inorganic N (DIN) concentration ($p = 0.77$); this indicates that DIN loads are controlled mainly by climatic and orographic factors that affect the amount of precipitation.

Mean annual NO_3^- concentrations in precipitation ranged about two-fold, from $9.7\,\mu mol\,l^{-1}$ at Brooklyn Lake to $21.1\,\mu mol\,l^{-1}$ at Manitou (Table 1), whereas mean annual NH_4^+ concentrations ranged more than those of NO_3^-, from $4.2\,\mu mol\,l^{-1}$ at Molas Pass to $16.9\,\mu mol\,l^{-1}$ at Alamosa. The four sites with the highest NH_4^+ concentrations were among the six easternmost sites, which are closest to agricultural sources on the plains and to transportation sources in the Denver–Boulder–Fort Collins urban corridor. The % of wet N deposition that consisted of NH_4^+ (on a molar basis) at these 15 sites, ranged from 28% at Molas Pass to 55% at Alamosa. The mean annual value for all 15 NADP sites was 37%.

Concentrations of NO_3^- and NH_4^+ in snowpack are generally consistent with those measured in wet deposition at nearby NADP sites (Turk et al., 2001). The mean NO_3^- concentration (1993–97) at the 27 high elevation sites sampled annually was $9.6\,\mu mol\,l^{-1}$, and the mean NH_4^+ concentration was $3.9\,\mu mol\,l^{-1}$. The highest NO_3^- concentrations were found at sampling locations near the Buffalo Pass and Dry Lake NADP sites in northern Colorado, locations at which sulfur isotope data suggests that local power plants contribute to atmospheric deposition (Mast et al., 2001).

3.2. Concentrations and loads of inorganic nitrogen in dry deposition

Dry deposition is generally more difficult and expensive to measure than wet deposition and thus, is measured at fewer locations. Additionally, there is no consensus as to the best method of measuring dry deposition. There are three CASTNET sites within the study region, however, that have estimated dry deposition rates of inorganic N for several years using consistent methods, which should allow the detection of time trends. These sites are located at elevations of 2743 m in Rocky Mountain National Park, 2926 m near Gunnison, CO (Gothic site), and 3178 m at the

Table 2
Mean annual dry deposition of N at three Clean Air Status and Trends Network (CASTNET) sites in the Rocky Mountains of Colorado and southern Wyoming

Site	Years	Mean dry deposition (kg N ha^{-1} yr^{-1})	S.D.
Gothic	1991–99	0.5	0.09
Rocky Mountain National Park	1995–98	1.4	0.11
Centennial	1992–99	1.1	0.09

GLEES (Centennial site) in Wyoming. The mean annual inorganic N loads at these sites range from 0.5 kg N ha^{-1} yr^{-1} at the Gothic site to 1.4 kg N ha^{-1} yr^{-1} at the Rocky Mountain National Park site (Table 2). Comparison of these values with those at nearby NADP wet-deposition sites indicates that dry deposition generally constitutes 25% to 30% of total atmospheric N deposition at these three sites. Zeller et al. (2000) similarly found that dry deposition of N averaged 30% of total atmospheric N deposition at the GLEES site during 1989–94.

Sievering et al. (1989) used a throughfall—incident approach to estimate a median dry plus fog deposition rate of 0.5–1 mg N m^{-2} d^{-1} during the 1987–88 growing seasons in a lodgepole pine canopy at 3100 m near Niwot Ridge. The method used included needle washing of natural and synthetic branchlets and a multiple regression approach to estimate canopy exchange of N. This growing season rate is equivalent to a load of 1.8–3.6 kg N ha^{-1} yr^{-1} if distributed over the entire year, however, subsequent studies (Sievering et al., 1992, 1996) found growing-season rates of dry deposition of N to be more than twice those of the dormant season in this area. Cress et al. (1995) found a rapid increase in dry deposition of N species in mid-April with the onset of upslope winds, which explains the greater rate of dry deposition during the growing season than the dormant season.

Sievering et al. (1992) estimated a dry-deposition rate of 0.7–0.9 kg N ha^{-1} yr^{-1} extrapolated to the alpine tundra on Niwot Ridge at 3525 m, but based on previously reported concentrations of five N species collected in ambient air during 1979–84 from a tower at 3050 m (Fehsenfeld, 1986; Fahey et al., 1986; Parrish et al., 1986; Roberts et al., 1988; Langford and Fehsenfeld, 1992) in a clearing near the lodgepole pine canopy that was previously sampled for throughfall at this site. Later work showed different ambient air concentrations on Niwot Ridge and the lower elevation sampling site resulting from different exposure to wind and weather patterns (Rusch and Sievering, 1995), suggesting that the previous extrapolation (Sievering et al., 1992) may have been invalid. Sievering et al. (1996) later reported a mean annual dry deposition rate of 2.8 kg N ha^{-1} yr^{-1} for 1993–94 based on concentrations of N species in ambient air at 3540 m on Niwot Ridge. This value was updated to 2.9 ± 0.8 kg N ha^{-1} yr^{-1} for 1991–95 based on data from the same ridgetop sampling location; this is the highest value reported for the Colorado Rockies (Sievering, 2001).

The mean annual-adjusted (Williams et al., 1998) wet-deposition rate for 1993–94 at the nearby Niwot Saddle NADP site was 3.2 kg N ha^{-1} yr^{-1}; therefore, the Sievering et al. (1996) mean annual dry-deposition estimate represented 47% of total N deposition, a value considerably greater than the % of total N deposition that consists of dry deposition at the CASTNET sites. Most of the difference in dry-deposition loads between the 1992 and 1996, 2001 studies of Sievering can be attributed to increased concentrations of N species in ambient air at Niwot Ridge from the 1979–84 data used in the 1992 paper to the data from the early 1990s used in the 1996 and 2001 papers. Sievering et al. (1996) showed that ambient concentrations of N-species in air at Niwot Ridge doubled from the mid-1980s to the mid-1990s; therefore, rates of dry-deposition of N probably increased similarly over this period.

Arthur and Fahey (1993) used throughfall measurements in a subalpine spruce-fir forest at the Loch Vale watershed to estimate that 56% of total atmospheric N deposition consisted of dry deposition during May through October 1986–87. Annual loads of N in dry deposition in this subalpine forest are likely to constitute less than 56% of total N deposition, however, in light of the previously discussed finding from Niwot Ridge that dry deposition in the growing season is about twice that during the dormant season.

Cloud and fog deposition have not been frequently measured in the Rocky Mountains of Colorado and southern Wyoming, but may significantly enhance reported rates of atmospheric N deposition. Sievering et al. (1989) noted that dew-wetted alpine tundra is occasionally observed at Niwot Ridge, but in a later study recorded no cloud or fog deposition events at Niwot Ridge during 1993–94 (Sievering et al., 1996). Nitrate and NH_4^+ concentrations $>50\,\mu mol\,l^{-1}$ have been measured in rime ice that forms in areas of alpine krummholz in Colorado and Wyoming (Borys et al., 1988; Lokupitiya et al., 2000). These concentrations of N species are four- to eight-fold greater than those of nearby snow deposition indicating that total atmospheric N loads are greater in areas where rime ice commonly forms.

Together, the data from the NADP and CASTNET networks and from the published studies discussed previously indicate that the total rate of atmospheric DIN deposition to the Colorado Rocky Mountains ranges from about 1 kg N ha^{-1} yr^{-1} in dry intermontane areas in southern Colorado (Alamosa) to about 7 kg N ha^{-1} yr^{-1} in high elevation areas of the Front Range east of the Continental Divide (Niwot Ridge). Total DIN deposition averages about 4.4 kg N ha^{-1} yr^{-1} at Loch Vale in Rocky Mountain National Park based on NADP and CASTNET data. Total DIN deposition may be even greater in localized areas that receive significant cloud, fog, or rime ice deposition.

3.3. *Organic nitrogen in atmospheric deposition*

Organic N is not measured in wet deposition at the NADP sites, nor in dry deposition at the CASTNET sites, but the relative contribution of organic N to total N loads in atmospheric wet deposition at the Green Lakes Valley in the Front Range during 1996–98 was 16% (Williams et al., 2001). Data from the Sierra Nevada suggest that organic N loads in wet deposition from drier regions of the west, such as central and western Colorado may represent as much as 25% of the total N load (Sickman et al., 2001). Thus, estimated total atmospheric N loads are probably 15–25% higher than inorganic N loads throughout the Rocky Mountain region of Colorado and southern Wyoming. Recent studies in Colorado that demonstrate direct uptake of amino acids in a non-mycorrhizal state by *Kobresia myosuroides*, a common alpine sedge, and its ability to compete with soil microbes for an amino acid, provide evidence of the importance of organic N in alpine ecosystems (Raab et al., 1996; Lipson and Monson, 1998), and suggest the value of more widespread measurements of organic N in the Colorado Rockies.

3.4. *Trend analysis*

Time trends in wet deposition of N were identified through Seasonal Kendall tests of volume-weighted mean monthly concentrations and loads in weekly samples from the beginning of the record at each site through December 2000. Correlation of concentration with precipitation amount was accounted for in the analysis through linear regression; therefore, only the residuals of these relations were used to identify time trends. Similar correlations were not explored between N load and precipitation amount because the two quantities are not independent. Most of the records from the 15 NADP sites extend back to the mid-1980s, but two—Brooklyn Lake and Wolf Creek Pass—began in 1992.

An analysis of trends in mean annual N loads in wet deposition at these 15 sites indicates statistically significant ($p<0.05$) trends of increasing N deposition at 5 of the sites—Buffalo Pass, Four Mile Park, Loch Vale, Niwot Saddle, and Snowy Range (Table 3). All

Table 3
Results of Seasonal Kendall trend analyses of NO_3^- concentrations, NH_4^+ concentrations, and N loads in wet deposition at NADP sites in the Rocky Mountains of Colorado and southern Wyoming

Site	Length of record	NO_3^- conc. (μmol l^{-1} yr^{-1})		NH_4^+ conc. (μmol l^{-1} yr^{-1})		N load (kg ha^{-1} yr^{-1})	
		Trend	*p*	Trend	*p*	Trend	*p*
East of Divide							
Alamosa	20 yr, 8 m	0.010	0.953	0.000	0.976	−0.001	0.173
Beaver Meadows	20 yr, 7 m	−0.052	0.513	0.058	0.363	0.001	0.204
Brooklyn Lake	8 yr, 3 m	−0.027	1.000	0.025	0.917	−0.001	0.835
Loch Vale	17 yr, 4 m	0.107	0.206	0.123*	0.022	0.004*	0.018
Manitou	22 yr, 2 m	−0.046	0.638	0.098	0.196	0.000	0.876
Niwot Saddle	16 yr, 6 m	0.316*	0.025	0.320*	0.002	0.013*	0.001
Snowy Range	14 yr, 8 m	0.192	0.062	0.169	0.075	0.005*	0.004
Sugarloaf	14 yr, 1 m	0.028	0.799	0.213	0.139	0.000	0.836
At Divide							
Buffalo Pass	16 yr, 9 m	0.373*	<0.001	0.284*	<0.001	0.010*	<0.001
Wolf Creek Pass	8 yr, 7 m	0.105	0.326	0.040	0.877	0.001	0.882
West of Divide							
Dry Lake	14 yr, 2 m	0.186	0.106	0.090	0.128	0.005	0.058
Four Mile Pk	13 yr, 0 m	0.197	0.168	0.258*	0.021	0.004*	0.004
Molas Pass	14 yr, 5 m	0.201	0.055	0.001	1.000	0.002	0.354
Sand Spring	21 yr, 9 m	0.003	0.958	−0.035	0.363	0.000	0.859
Sunlight Peak	12 yr, 11 m	0.219	0.268	0.117	0.220	0.003	0.080

*Trend is significant at $p<0.05$.

five of these sites have data since 1983–87, and four of the sites are at high elevation (>3000 m); the fifth (Four Mile Park) is at 2502 m. The slopes of the regression relations for these five sites indicate an annual increase that ranges from about 0.004 kg N ha^{-1} yr^{-1} at Four Mile Park to 0.013 kg N ha^{-1} yr^{-1} at Niwot Saddle. Four of the five sites (except Snowy Range) also showed significant increasing trends in NH_4^+ concentrations, but only two of these sites—Buffalo Pass and Niwot Saddle—had significantly increasing NO_3^- concentrations. These two sites also were the only ones with increasing trends in precipitation amount (data not shown). This suggests that the increasing N loads in wet deposition at Buffalo Pass and Niwot Saddle result from increasing NO_3^- and NH_4^+ concentrations as well as increasing precipitation amount, whereas the increasing N loads in wet deposition at Four Mile Park and Loch Vale result primarily from increasing NH_4^+ concentrations. The trend of increasing N load at the Snowy Range site is attributed to a combination of nearly significant increases in NO_3^- and NH_4^+ concentrations in wet deposition ($p = 0.06$ and 0.08, respectively).

Baron et al. (2000) used two-stage linear regression of log transformed data through 1999 and found significant increasing trends of NO_3^- concentrations (0.09 μmol l^{-1} yr^{-1}) at the Niwot Saddle NADP site and in NH_4^+ concentrations at the Loch Vale and Niwot Saddle sites (0.18 and 0.26 μmol l^{-1} yr^{-1}, respectively). An increasing trend in NO_3^- concentrations at Niwot Saddle was also found in the current study, but the magnitude is more than three-fold less than the trend reported by Baron et al. (2000) (Table 3). However, the trends in NH_4^+ concentrations in the current study are similar in magnitude and direction to those reported by Baron et al. (2000).

A significant increasing trend in wet deposition of 0.32 kg N ha^{-1} yr^{-1} at the Niwot Saddle site was previously reported for 1984–96 (Williams and Tonnessen, 2000), and a similar trend of increasing wet deposition of 0.35 kg N ha^{-1} yr^{-1} was reported at the Snowy Range site for 1986–90 (Williams et al., 1996a). About half of the 1984–93 increase at Niwot Saddle was attributed to an increase in precipitation amount, and about half to an increase in the volume-weighted concentration of NO_3^- (Williams et al., 1996a). The trends reported here for Niwot Saddle and Snowy Range are consistent with the direction of trends reported by Williams et al. (1996a) and Williams and Tonnessen (2000), except that: (1) the trends reported here are more than an order of magnitude lower (Fig. 2, Table 3), and (2) the increasing NH_4^+ concentrations noted here contributed significantly to the increasing N loads. One likely reason for these differences in the slope of trends is that Williams and Tonnessen (2000) used linear regression analysis of annual N loads, whereas Seasonal Kendall analysis of monthly weighted mean values was used in the present study. The nonparametric analysis used here is preferred to linear regression because these chemical data fail the standard Wilk-Shapiro test (Shapiro and Wilk, 1965) for normality; therefore, linear regression is not considered a valid approach to identify monotonic time trends (Helsel and Hirsch, 1992). The trend of increasing N loads in wet deposition at Niwot Saddle identified by Seasonal Kendall analysis indicates that about 50% of the increase results from increasing precipitation, about 25% from increasing NO_3^- concentrations, and about 25% from increasing NH_4^+ concentrations.

The Niwot Ridge site showed an increase in annual precipitation amount and a decrease in mean annual air temperature from 1951 to 95 (Williams et al., 1996b). The results obtained here concur that precipitation amount at Niwot Saddle increased significantly from 1984 to 2000, but found only one other NADP site in the region with an increasing trend, suggesting the lack of a region-wide increase in precipitation. The decrease in air temperature is counter to the general predicted effects of climatic warming driven by increasing concentrations of CO_2 in the atmosphere, and no consensus has been reached that colder and snowier conditions should be expected at high elevations in the Rockies in the future as a result of predicted climate change (Hauer et al., 1997).

The Gothic and Centennial CASTNET sites show no statistically significant trend in annual dry N deposition over their periods of record, which extend back to the early 1990s. The site at Rocky Mountain National Park was not analyzed for time trends because the record was shorter than the 8-year minimum considered necessary for Seasonal Kendall analysis (Helsel and Hirsch, 1992). A measurement site at Niwot Ridge showed more than 20-fold greater atmospheric concentrations of some N species than those measured in remote unpolluted air (Fahey et al., 1986), and a more recent study has shown that the air concentration of N species at Niwot Ridge during the growing season more than doubled from the mid-1980s to the mid-1990s (Sievering et al., 1996).

Surprisingly, the monthly mean concentration and monthly flux of particulate NO_3^- and NH_4^+ in dry deposition have decreased significantly through the 1990s at the Gothic and Centennial CASTNET sites (the decreased NO_3^- flux at the Centennial site was of marginal statistical significance, $p = 0.10$). These data are inconsistent with the observations from Niwot Ridge, but the difference can probably be attributed to the different geographic location and sample periods at these sites.

In conclusion, the rate of wet deposition of N increased from the 1980s to 2000 at 5 of 15 NADP sites across the Colorado and southern Wyoming Rockies, but no change in N deposition is evident at the 10 other

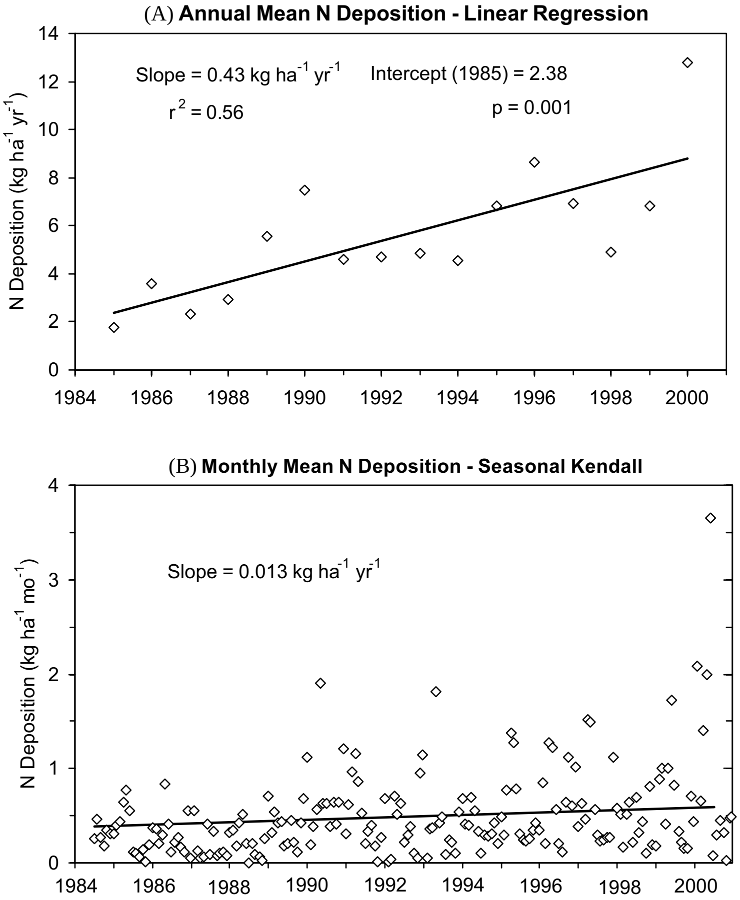

Fig. 2. Trends in wet deposition of atmospheric N at the Niwot Saddle NADP site: (A) Linear regression of mean annual values and (B) seasonal Kendall analysis of monthly volume-weighted values (site location shown in Fig. 1).

NADP sites in this region. Additionally, no significant trend in dry deposition of N since the early 1990s is supported by data from two CASTNET sites, although the paucity of data collection sites and short time records for dry deposition make a regional trend difficult to recognize. Data from Niwot Ridge support an increasing trend of N species concentrations in air from the 1980s to the 1990s, but rates of dry deposition have not been consistently estimated from these data. The Niwot Ridge site also had the greatest increase in wet deposition of N from the mid-1980s through 2000. All three high-elevation (>3000 m) NADP sites in the Front Range with data since the 1980s show significant increases in N loads in wet deposition. The two NADP sites (Beaver Meadows and Sugarloaf) at lower elevations ($\cong 2500$ m) in the Front Range, however, showed no significant trend in wet deposition of N during a similar time interval. The reason for a lack of trends at lower elevation sites in the Front Range is unclear. Overall, these data do not indicate a widespread increase in atmospheric N deposition throughout the Colorado Rockies from the mid-1980s to the 1990s, but imply such an increase only at high elevation in the Front Range. These data also suggest a need to re-calculate these trends periodically because two sites (Dry Lake and Sunlight Peak) that did not show a significant trend in wet deposition of N, show an increasing trend of borderline statistical significance ($0.05<p<0.10$); this suggests that a continuation of present patterns may result in significant increasing trends within the next few years.

3.5. *Origin of N in atmospheric deposition—East or West?*

The Colorado Rocky Mountains are in an area of predominantly west wind; this suggests that most sources of airborne N should originate from west of the Rocky Mountains (Hansen et al., 1978; Sievering et al., 1996). Several investigators have reported, however, that significant amounts of atmospheric N deposition—especially in the Front Range—originate from the east (Langford and Fehsenfeld, 1992; Baron and Denning, 1993; Heuer et al., 2000). Additionally, Parrish et al. (1990) inferred from N species concentrations and source patterns in air at Niwot Ridge that N from sources to the east can become mixed into high-level flow from the west, and later be deposited from these eastward flowing air masses. The data analyzed from NADP sites are broadly consistent with a source of at least some of the NH_4^+ in wet atmospheric deposition from the east because the four sites with the highest mean concentrations of NH_4^+ were among the six easternmost sites in the region.

The relative amount of atmospheric N deposition that originates from east of the Front Range in the Denver–Boulder–Fort Collins urban corridor varies with season. Precipitation samples collected at NADP sites west of the Continental Divide during 1992–97 and 1999 had lower mean annual volume-weighted concentrations of NO_3^- and NH_4^+ than samples collected east of the divide (Heuer et al., 2000; Baron et al., 2000), and precipitation samples collected during the summer (May–September) of 1992–97 showed a similar pattern (Heuer et al., 2000). In contrast, snow samples collected west of the Continental Divide during winter-pack surveys had higher NO_3^- concentrations than samples from east of the Divide, but NH_4^+ concentrations showed no east–west difference (Heuer et al., 2000). These results are consistent with a large contribution of N to atmospheric deposition in summer from the Denver–Boulder–Fort Collins urban corridor associated with differential heating of air at low elevations that moves upvalley toward the mountains. These upslope events are commonly associated with convective thunderstorms (Toth and Johnson, 1985). Baron and Denning (1993) found that a significant amount of the NO_3^- and NH_4^+ measured in precipitation samples at the Beaver Meadows and Loch Vale NADP sites in the Rocky Mountain National Park originated east of the mountains. The lower elevation Beaver Meadows site received N deposition from the east throughout the year, whereas the higher elevation Loch Vale site received N deposition from the east only during summer.

One region, the Yampa River Valley, does not follow the typical east–west deposition pattern observed in the Colorado Rockies. The concentrations of NO_3^- in winter snowpack samples from the Yampa River Valley are the highest measured anywhere in the Colorado Rocky Mountain region (Turk et al., 1992, 2001). This is consistent with the moderately high mean annual wet-deposition loads (1995–99) of 2.5 kg N ha^{-1} yr^{-1} at Dry Lake and 3.3 kg N ha^{-1} yr^{-1} at Buffalo Pass in this Valley (Table 1). This region is downwind (east) of two large power plants in northwestern Colorado whose emissions exceed 5000 t yr^{-1} of NO_x (Dickson et al., 1994), and sulfur isotope data suggests that these power plants contribute significantly to atmospheric sulfur deposition at downwind locations. The large annual N loads in wet deposition at Dry Lake and Buffalo Pass, which are unusual for mountainous areas west of the Continental Divide in Colorado, have been attributed to the power plant emissions (Turk and Campbell, 1997).

Recent data collected near Telluride suggests that southwest Colorado also receives elevated levels of atmospheric N deposition that may be influenced by emissions from six power plants located near the Four Corners (Williams and Manthorne, 2001). Emissions from these power plants may also contribute to atmospheric N loads at the Molas Pass and Wolf Creek Pass NADP sites.

4. Summary

Atmospheric N deposition in the Rocky Mountain Region of Colorado and southern Wyoming ranges from 1 to 7 kg N ha^{-1} yr^{-1}, and may be even greater at high elevations (>3500 m) in the Front Range. From 25% to 30% of total inorganic N deposition consists of dry deposition, but these values are less certain than those of wet deposition because fewer measurements are available, and the measurements are less accurate than those used for wet deposition. Atmospheric N deposition is generally greater east of the Continental Divide than west of the Divide in the Front Range, except in areas that are directly downwind of large power plants, such as the Buffalo Pass NADP site. Despite the prevailing west winds at this latitude, westward upslope movement of polluted air masses from the Denver–Boulder–Fort Collins metropolitan area appears to be contributing to atmospheric N, particularly during summer. A tendency toward higher NH_4^+ concentrations in wet deposition at NADP sites east of the Divide than those west of the Divide is evident and probably reflects agricultural and vehicle sources of atmospheric NH_4^+ from east of the Rockies.

Five of 15 NADP sites show trends of increasing wet deposition of N from the mid-1980s to 2000. A region-wide trend of increasing wet deposition of N is not evident, but three of the five sites with increasing trends, Loch Vale, Niwot Saddle, and Snowy Range, are at high elevations east of the Divide in the Front Range. The trend at the Loch Vale site is attributed mainly to increasing NH_4^+ concentrations in precipitation, whereas the trend at the Niwot Saddle site results equally from increasing precipitation and increasing concentrations of NO_3^- and NH_4^+ in precipitation. The increasing trends of wet deposition of N at the Niwot Saddle and the Snowy Range sites are more than an order of magnitude less than reported in earlier studies, because those studies erroneously used linear regression to examine trends in data that are not normally distributed.

The current upward trends in population growth and energy use in Colorado and the west indicate that long-term monitoring of the chemistry of atmospheric deposition is needed to contribute to continued assessments of the effects of atmospheric N deposition on high elevation ecosystems. Given the upward trends in NH_4^+ concentrations at high elevation in the Front Range, the role of N from agriculture cannot be ignored in such assessments. Data from long-term measurement sites that are located close to monitoring and research on water chemistry and ecosystem processes such as Loch Vale, Niwot Ridge, and GLEES are particularly valuable in assessing trends and effects, and warrant the highest priority for funding.

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