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CRITICAL LOADS FOR INORGANIC NITROGEN DEPOSITION IN THE COLORADO FRONT RANGE, USA

Mark W. Williams
Department of Geography and
Institute of Arctic and Alpine Research,
University of Colorado, Boulder

Kathy A. Tonnessen
Air Resources Division
National Park Service
Denver, CO

Corresponding Address†

Mark W. Williams
INSTAAR and Dept. of Geography
Campus Box 450
Boulder, CO 80309-0450
Telephone: 303/492-8830
Internet: markw@snobear.colorado.edu

ABSTRACT

We suggest an empirical approach for determining critical loads for inorganic N deposition in wetfall to the central Rocky Mountains. We define "critical loads" as a deposition amount above which natural resources can be negatively affected. The arithmetic average from 1992 to 1996 of annual inorganic N deposition in wetfall at the eight NADP sites located at elevations greater than 2,500 m in the central Rocky Mountains ranged from 2.5 to 3.5 kg ha^{-1} yr^{-1}. In contrast, inorganic N deposition was less than 2.5 kg ha^{-1} yr^{-1} at all 23 NADP sites less than 2,500 m in elevation. At the Niwot Ridge NADP site in the Colorado Front Range, a simple linear regression of inorganic N in wetfall with time shows a significant increase in deposition of inorganic N in wetfall at the rate of of 0.32 kg ha^{-1} yr^{-1} ($r^2 = 0.62$; $p < 0.001$, $n = 13$). In turn, the increasing amount of inorganic N in wetfall is causing episodic acidification in headwater catchments of the Green Lakes Valley in the Colorado Front Range, with acid neutralizing capacity (ANC) values below 0 μeq/L in surface waters during snowmelt runoff at 9-ha and 42-ha sampling sites. At present rates of ANC decrease, we can expect the 9-ha and 42-ha sites to become chronically acidified within the next decade and the 220-ha basin of Green Lake 4 to become episodically acidified. A synoptic survey in 1995 of 91 high-elevation lakes in the central Rocky Mountains suggests that water quality is being affected by inorganic N in wetfall throughout the region. Federal land managers are required to "err on the side of protection" when assessing the amount of deposition that will alter ecosystem processes. However, given the political and economic ramifications of policy decisions, land managers are aware of the need to provide a scientific basis for these decisions and to balance conflicting needs. To achieve this balance and to allow for natural resource protection, we make a conservative recommendation that critical loads of inorganic N in wetfall to Class 1 areas in in the central Rocky Mountains be set at 4 kg ha^{-1} yr^{-1}. Target loads my be set at lower levels of inorganic N deposition in wetfall to allow a margin of safety to protect extremely sensitive natural resources.

Key words: nitrogen, critical loads, acidification, Rocky Mountains, alpine, lakes, streams, national parks, wilderness areas

Key Phrases: critical loads for annual nitrogen deposition in wetfall to the Rocky Mountains; acidification of surface waters from nitrogen deposition.

INTRODUCTION

Critical loads have been widely accepted in Europe and Canada as a basis for negotiating control strategies for transboundary air pollution [Posch et al., 1997]. A workshop on critical loads held under the auspices of the United Nations Economic Commission for Europe (UN/ECE) in 1988 provided the first working definition of critical loads as "the quantitative estimate of an exposure to one or more pollutants below which harmful effects on specified sensitive elements of the environment do not occur according to present knowledge" [Nilsson and Grennfelt, 1988]. The scientific basis for calculating and mapping critical loads in Europe has developed sufficient credibility such that sulfur emissions were limited by the Second Sulfur Protocol [UN/ECE, 1994].

In both European countries and North America, attention has recently shifted from ecosystem damage caused by sulfur deposition to potential ecosystem damage caused by inorganic nitrogen (N) deposition. In some European forests, chronically high N deposition has exceeded the assimilation capacity of ecosystems, leading to the release of nitrate (NO_3^-) from terrestrial ecosystems into surface waters [Dise and Wright, 1995]. These ecosystems have changed from N-limited to no longer N-limited systems, an ecosystem condition sometimes referred to as "nitrogen saturation" [Ȧgren, 1983; Aber et al., 1989], which has been linked to forest decline [Schulze, 1989] and acidification of soils and surface waters in Europe [Durka et al., 1994]. Similarly, in northeastern North America, there is concern that present levels of N deposition are causing perturbations in the N cycle and in ecosystem function [Aber, 1992; Stoddard, 1994]. Nitrogen saturation of forested catchments has contributed to environmental problems such as reduced drinking water quality, NO_3^--induced toxic effects on freshwater biota, disruption of nutrient cycling, increased soil acidification and aluminum mobility, and increased emissions from soil of nitrogeneous greenhouse gases [Fenn et al., 1998].

Current levels of atmospheric deposition of N have been linked to changes in the ecosystem function of high-elevation catchments in the central Rocky Mountains of North America. In general, water quality of lakes in the Rocky Mountains are pristine, with the median value of NO_3^-

concentrations less than 1 μeq/L [Psenner, 1989]. However, these high-elevation ecosystems are relatively sensitive to changes in the flux of energy, chemicals and water compared to downstream ecosystems, because of extensive areas of exposed and unreactive bedrock, rapid hydrologic flushing rates during snowmelt, limited extent of vegetation and soils, and short growing seasons [Williams et al., 1993; NAPAP, 1998]. Hence, small changes in atmospheric deposition have the potential to result in significant changes in ecosystem dynamics and water quality [Williams et al., 1996a]. Ambient concentrations of anthropogenically-fixed N measured in the early 1980's at Niwot Ridge in the Colorado Front Range were 30-fold greater than pre-industrial levels [Fahey et al., 1986] and attributed to fossil fuel combustion [Lewis et al., 1984]. Additional measurements of ambient N at Niwot Ridge in the early 1990's have shown that anthropogenically-fixed N in the atmosphere has since doubled [Rusch and Sievering, 1995]. In response to atmospheric deposition of anthropogenically-fixed N, some catchments of the Front Range of the Rocky Mountains have shifted from N-limited ecosystems to N-saturated ecosystems (Williams et al., 1996b; NAPAP, 1998).

At present, critical loads for N deposition have not been established for the United States. The Clean Air Act Amendments (CAAA) of 1990, section 404, called for the Environmental Protection Agency (EPA) to prepare a report on the feasibility and the environmental effectiveness of setting an acid deposition standard to protect sensitive aquatic and terrestrial resources. The completed report includes a number of modeling analyses that project the effect of reductions in both S and N deposition in areas well-studied during the National Acid Precipitation Assessment Program (NAPAP, 1995). The conclusions of the EPA's analysis are that: (1) the uncertainties associated with effects of N on ecosystems are such that critical loads cannot be set at this time, (2) there had been no policy decision made regarding the level of acceptable damage to systems, and (3) that any critical load standards would have to be set on a regional basis and then enforced with regional pollution abatement strategies. Fenn et al. [1998] have documented the ecosystem problems that are presently occurring in North America as a result of excess N in atmospheric deposition, including high-elevation areas of the Rocky Mountains.

However, the authors were unable to recommend practical strategies to define critical loads for N deposition.

There is some urgency in defining critical loads for N deposition in the western United States. A recent analysis of national patterns in atmospheric deposition of N showed that 45 of 217 sites demonstrated an increasing trend in N deposition. More than half of these sites were in remote areas previously thought to be relatively pristine, including Rocky Mountain National Park in Colorado, Bryce Canyon in Utah, and Sequoia National Park in California [Gosz and Murdoch, 1998]. Aber et al. [1998] recently evaluated the response of the US regulatory community to atmospheric deposition of pollutants as follows: "Since the passage of the 1990 CAAA, the US regulatory community has not supported substantial, additional research on acidic deposition. Unlike the European Community, which has pursued an active and well-coordinated international program on the effects of N deposition [the NITREX program: Wright and van Breeman 1995; Wright and Rasmussen, 1998], and on critical loads for N [Nilsson and Grennfelt, 1988; Henriksen et al., 1992; Warfvinge and Sverdrup, 1992; Wright and Rasmussen, 1998], research on this topic in the United States remains scattered and piecemeal. Policy and regulatory activity have also, until very recently, been virtually nonexistent."

We suggest an empirical approach for setting critical loads for N deposition in the central Rocky Mountains. Here we define "critical loads" as a deposition amount above which natural resources can be negatively affected. To accomplish this task, we use published and original data to: (1) estimate emissions and deposition amounts of inorganic N in and near the central Rocky Mountains; (2) use intensive data from the well-studied Green Lakes Valley of the Colorado Front Range to determine hydrochemical changes in surface waters at present deposition levels of inorganic N in wetfall; (3) use synoptic data to evaluate the spatial extent of N leakage in the Colorado Front Range; (4) evaluate current and potential changes in ecosystem function as a result of inorganic N in atmospheric deposition; and (5) use this information to recommend critical loads for inorganic N deposition to high-elevation catchments of the central Rocky Mountains. While we concentrate here on a specific region in the Rocky Mountains because of data

availability, this analysis provides insights into ecological risk assessment and management responses relevant to mid-latitude montane regions throughout the world.

SITE DESCRIPTION

Nitrogen Emissions and Deposition

Nitrogen emissions from stationary and mobile sources and deposition in wetfall were evaluated for a five-state region that includes the central Rocky Mountains and much of the intermountain West: Wyoming, Colorado, Utah, Arizona and New Mexico (Figure 1). Water quality samples were collected throughout the Colorado Front Range, which we define as the region from the Arkansas River north along the Continental Divide through Rocky Mountain National Park to the Colorado-Wyoming Border [Arno and Hammerly, 1984]. The Colorado Front Range rises directly from the Denver-Colorado Springs-Fort Collins metropolitan area. This geographical setting results in high-elevation basins of this portion of the Continental Divide being located just west of large urban areas and agricultural centers. Topographically, Pleistocene glaciation has resulted in high-elevation areas of the eastern slope of the Colorado Front Range generally having deeper and steeper canyons with less soil development compared to high-elevation areas to the west of the Continental Divide.

Water Quality: Intensive Site

Intensive samples were collected for wetfall and surface water chemistry in the Green Lakes Valley (40 03' N, 105 35' W) of the Colorado Front Range (Figure 1). The Green Lakes Valley is an east-facing headwater catchment that abuts the Continental Divide and is located entirely within the Arapaho-Roosevelt National Forest. The basin is 700 ha in area and ranges in elevation from 3,250 m to ≈ 4,000 m (Figure 1). The catchment appears typical of the high-elevation environment of the Colorado Front Range, and includes Niwot Ridge, where research has been conducted since the early 1950's [Caine and Thurman, 1990]. Bedrock is crystalline, with about 80% of the basin composed of exposed bedrock and talus.

The catchment is a linear cascade of five lakes located on the hydrologic axis of the basin, with seven sites sampled about weekly for water quality and one site (outlet of Green Lake 4)

gauged for continuous discharge measurements (Figure 1). Here we present results from three of the sampling sites. The stream at Arikaree cirque (ARIK) drains a 9-ha area of ice and snow at 3,785 m; composed of the stagnant Arikaree Glacier and surrounding rock and boulder slopes with negligible vegetation cover. The downstream Navajo (NAV) sampling site is 42 ha in area at an elevation of 3,700 m. The outflow from Green Lake 4 (GL4) drains an area of 220 ha at an elevation of 3,550 m, with Green Lake 5 located between the NAV and GL4 sampling sites. About 80% of the annual precipitation in the Green Lakes Valley occurs as snow. Streamflows are markedly seasonal, varying from less than 0.1 $m^3 s^{-1}$ during the winter months to greater than 1.5 $m^3 s^{-1}$ at maximum discharge during snowmelt just below Lake Albion at the lower end of the valley [Caine, 1996]. Surface waters are dilute, with acid neutralizing capacities (ANC) generally less than 200 μeq/L at all sampling sites [Caine and Thurman, 1990].

Niwot Ridge forms the northern boundary of Green Lakes Valley (Figure 1) and is an UNESCO Biosphere Reserve and a Long-Term Ecological Research (LTER) network site. The LTER network site is a participant in the National Atmospheric Deposition Program (NADP) and has maintained an NADP wet deposition collector since 1984 on the Niwot Ridge saddle at an elevation of 3,500 m (Figure 1).

Water Quality: Synoptic Lake Samples

Synoptic samples for water quality were collected in 1995 from lake outlets throughout high-elevation Wilderness Areas of the Colorado Front Range. Special emphasis was placed on sampling Wilderness Areas because of the specific mandates that the CAAA provides for Federal Land Managers to protect air quality in Class I areas. Four subregions were sampled (from south to north): Sangre de Cristos, Mount Evans, Indian Peaks, and Rawah Wilderness Areas (Figure 1). For comparison purposes, we also collected samples from lake outlets in three Wilderness Areas located to the west of the Continental Divide in Colorado: Eagles Nest, Holy Cross, and Weinimuche Wilderness Areas. Lakes were sampled in cooperation with the Arapaho and Roosevelt National Forests and the Rocky Mountain Forest and Range Experiment Station

[Musselman et al., 1996].

METHODS

Nitrogen Emissions and Deposition

Point and area sources of nitrogen emissions were compiled for the 11 western states for 1990 by the National Acid Precipitation Assessment Program [Dickson et al., 1994]. Point sources were stationary sites that emitted more than 1,000 tons/year of NO_x. Area sources were compiled on a county-by-county basis within each state, and include stationary sources that emitted less than 1,000 tons/year of NO_x and mobile sources such as diesel- and gasoline-powered vehicles.

The NADP program operates about 200 wet precipitation collectors throughout the continental United States [NADP, 1984-1996]. NADP samples are collected and analyzed using the same protocols at all sites so that results among sites can be compared. Here we present results from all NADP sites in the five-state region of the intermountain West for the period 1992-1996. Additionally, we present the full record of inorganic N deposition in wetfall from the Niwot Ridge NADP collector. Previous research by Williams et al. [1998] has shown that blowing snow causes the NADP wet chemistry collector on Niwot Ridge to overestimate annual deposition of N in wetfall by about 30%; the NADP results for Niwot Ridge were adjusted to account for the over-collection problems caused by blowing snow.

Water Quality: Intensive and Synoptic Sites

Water quality samples were collected at both intensive and synoptic sites using the same sampling and analytical protocols. Water samples were collected as grab samples in polyethylene bottles soaked with DI water overnight and then rinsed copiously five times; bottles were further rinsed three times with sample water at the time of collection.

Water quality samples at GL4 were collected about weekly from the initiation of snowmelt to fall freeze-up and then about monthly, starting in 1984. Water quality samples at ARIK and NAV sites were collected about weekly from the initiation of snowmelt to fall freeze-up for each of those years. Samples were analyzed for all major solutes except NO_3^- and ammonium (NH_4^+); NO_3^- analysis started in 1985 and NH_4^+ analysis was initiated in 1993. Water samples were transported the day of collection 4 km to the Kiowa environmental chemistry laboratory and sample analysis was immediately initiated. Synoptic samples at 91 high-elevation lakes in Colorado were collected in 1995 at the peak of the growing season, from 25 July through 16 August. This time period was selected because inorganic N concentrations in surface waters are generally near or at their annual minima as a result of biological assimilation [Stoddard, 1994]. All water samples from the synoptic survey were collected at or near lake outlets and transported in coolers to the analytical laboratory.

All water samples were analyzed for pH, acid neutralizing capacity (ANC), conductance, major ions and reactive silicate (Si), following the protocol of Williams and Melack (1991a). Conductance, pH and ANC were measured immediately after transport to the wet chemistry laboratory; ANC was measured using the Gran titration technique. Subsamples were immediately filtered through pre-rinsed (300 ml), 47-mm Gelman A/E glass fiber filters with ca. 1-micron pore size. Filtered samples were stored in the dark at 4°C for subsequent analyses within one to four weeks; except for NH_4^+. Ammonium was determined colorimetrically within 24 hr of collection, on a Lachat flow injection analyzer using a phenolate reaction enhanced by nitroprusside; detection limit was 0.7 μeq/L and precision 2.7%. Anions were measured using ion chromatography (Dionex DX 500) employing chemical ion suppression and conductivity detection. The detection limit for NO_3^- was 0.1 μeq/L and precision was 1.5%.

RESULTS

Nitrogen Emissions and Deposition

In the Rocky Mountain region of the US, there are 58 point sources that produce more than 1,000 tons per year of NO_x emissions [Dickson et al., 1994] (Figure 2). Large emitters are located near both urban areas and in more remote areas where large coal-fired facilities have been sited. Point sources in remote areas include the coal-fired facilities in southern Wyoming (Bridger power plant), in western Colorado (Craig and Hayden power plants), and in the Four Corners area near Mesa Verde National Park. Its worth noting that in Colorado, there is a cluster of point emission sources just to the east of the Colorado Front Range that is associated with the Denver-Colorado Springs-Fort Collins metropolitan area.

Point sources of NO_x are not the only source of N emissions in this five state region. Counties with NO_x emissions greater than 5,000 tons per year were generally associated with urban centers that had populations greater than 50,000 (Figure 3). To illustrate for Colorado, county emission rates greater than 5,000 tons per year occurred in the Denver and Colorado Springs metropolitan areas to the east of the Rocky Mountains. Similarly in Arizona, emission rates of NO_x greater than 5,000 tons per year were estimated for sources near Phoenix and Tucson.

Nitrogen emissions from these five mountain states of 3,560 tons per day of NO_x were about the same as the 3,685 tons per day of NO_x emitted in California alone. Mobile sources-primarily diesel and gasoline vehicles-accounted for about 50% of all NO_x emissions in each state. To illustrate for Colorado, mobile sources of 390 tons per day of NO_x emissions accounted for 46% of the 830 tons per day of NO_x emitted throughout the state in 1990. For the 11 states as a whole, mobile sources accounted for 48.5% of the 17,500 tons per day of NO_x emissions.

It is also worth noting that many rural counties in Wyoming had emission rates between 1,000 and 5,000 tons per year (Figure 3). These NO_x emissions in Wyoming are not associated with either point sources or urban populations. These NO_x emissions in rural Wyoming are thought to be from oil and gas operations, which individually do not produce enough emissions

to be classified as point sources. It is important to note that in southwestern Wyoming the Bureau of Land Management is considering permits for development of a large natural gas field [BLM, 1996]. The NO_x emissions associated with this energy development might well have effects on N-loading to Wilderness Areas in both northern Colorado and southern Wyoming.

Annual deposition of inorganic N in wetfall ($NH_4^+ + NO_3^-$) from 1992 to 1996 was greater at NADP sites located in montane areas compared to surrounding lowlands (Figure 4). The arithmetic average of annual inorganic N deposition in wetfall at the eight NADP sites greater than 2,500 m in elevation ranged from 2.5 to 3.5 kg ha^{-1} yr^{-1}. In contrast, inorganic N deposition was less than 2.5 kg ha^{-1} yr^{-1} at all 23 NADP sites less than 2,500 m in elevation. The largest amounts of inorganic N in wetfall for all NADP sites in the five state region were at high-elevation sites located on the east side of the Colorado Front Range: Niwot Ridge, Loch Vale in Rocky Mountain National Park, and the Glacier Lakes, Wyoming site on the northern end of the Colorado Front Range.

The greater amounts of inorganic N deposition in montane areas of Rocky Mountain states compared to surrounding lowlands may be in part from orographic precipitation. Results from the 1994 NADP annual report are used to illustrate the increase in chemical loading that occurs with elevation in the Rocky Mountains and other high-elevation catchments (Figure 5). For this example we focus on NO_3^-, but other chemical species show a similar pattern. The Pawnee site is located about 50 km east of the Colorado Front Range in the Great Plains, at an elevation of 1,641 m. Beaver Meadows is a montane site located on the eastern edge of Rocky Mountain National Park at an elevation of 2,490 m. The Loch Vale site is located near the continental divide in Rocky Mountain National Park at an elevation of 3,159 m. Precipitation amount increases with elevation, from 24 cm at the Pawnee site to 117 cm at Loch Vale. In contrast, annual volume-weighted mean concentrations of NO_3^- in wetfall decrease with elevation, from 20.9 μeq/L at the Pawnee site to 14.3 μeq/L at Loch Vale. However, annual loading of NO_3^- increases with elevation because of the large increase in precipitation amount with elevation. The annual loading of 14.3 kg ha^{-1} of NO_3^- at Loch Vale was about four times the annual

loading of 3.1 kg ha^{-1} at the Pawnee site.

Annual deposition of inorganic N in wetfall at the Niwot Ridge NADP site from 1984 to 1996 about doubled from 1.95 kg ha^{-1} yr^{-1} for the four-year period from 1985 to 1988 to 3.75 kg ha^{-1} yr^{-1} for the four-year period from 1989 to 1992 (Figure 6a). Annual deposition of inorganic N in wetfall at the NADP site on Niwot Ridge has remained near or above 3.0 kg ha^{-1} yr^{-1} since 1992. A simple linear regression of inorganic N in wetfall with time shows a significant increase in deposition of inorganic N in wetfall at the rate of of 0.32 kg ha^{-1} yr^{-1} (r^2 = 0.62; $p < 0.001$, n = 13). Earlier and comparable measurements of annual NO_3^- deposition extend the record back to 1982 and suggest that the increase in deposition of inorganic N in wetfall began in the early 1980's [Caine and Reddy, 1988]. Previous analysis of the NADP record by Williams et al. [1996b] for a shorter time period shows that about half of the increase in N deposition is from increasing concentrations of N in wetfall and about half from increasing amounts of annual precipitation.

Water Quality: Intensive Site Study

The increased loading of inorganic N in wetfall appears to be causing changes in the hydrochemistry of surface waters in Green Lakes Valley. A time series of NO_3^- concentrations from the outlet of GL4 shows that maximum concentrations occur at the initiation of snowmelt runoff in each year (Figure 6c). These NO_3^- concentrations of about 30 μeq/L are three times the NO_3^- concentrations of about 10 μeq/L reported by Williams et al. [1996c] for the snowpack at maximum accumulation on Niwot Ridge. At the initiation of snowmelt each year, a large pulse of NO_3^- enters surface waters in the Green Lakes Valley. To place these NO_3^- values in context, the comparable Emerald Lake basin in the Sierra Nevada of California is not subject to elevated amounts of inorganic N in wetfall [Williams et al., 1995] and has an annual maximum concentration of about 12 μeq/L of NO_3^- during snowmelt runoff [Williams and Melack, 1991b]. These elevated values of NO_3^- during snowmelt runoff in Green Lakes Valley are consistent with Stage 1 of N saturation as defined by Stoddard [1994].

Perhaps more importantly, leaching of NO_3^- in surface waters of Green Lakes Valley is now occurring during the growing season in response to wetfall amounts of about 3 kg ha^{-1} yr^{-1} of inorganic N (Figure 6b). At the start of the water quality record in the mid-80's, NO_3^- concentrations on the receding limb of the hydrograph during the growing season declined to detection limits. Paralleling the increase in NO_3^- loading from wet deposition in the late 1980's, there was an increase in the annual minimum concentrations of NO_3^- during the summer in Green Lake 4, from below detection limits in 1985 to about 10 μeq/L in 1990. A linear regression analysis of the annual minimum concentrations of NO_3^-, with years 1989 and 1990 removed because they were more than 3 standard deviations above the mean, shows that annual minimum concentrations of NO_3^- in surface waters are increasing at the rate of 0.23 μeq/L per year (r^2 = 0.57, p = 0.005, n = 11). The trend of increasing values of NO_3^- in surface waters during the growing season are consistent with initiation of Stage 2 of N saturation as defined by Stoddard [1994] and with the definition of N saturation given by Fenn et al. [1998].

Apparently in response to the increased deposition of inorganic N in wetfall, ANC has been decreasing in GL4 since the mid-1980's. Here we present a trend analysis of ANC at GL4, based on the approach of Caine [1995] (Figure 7). A simple linear regression of ANC versus time shows that ANC in GL4 has been decreasing at the rate of 2.5 μeq/L per year. While the r^2 of 0.24 is low, the slope of -0.0069 is significant at the α = 0.05 level. The decrease in ANC at GL4 is coincident with the increase in deposition of inorganic N in wetfall.

Episodic acidification (ANC < 0 μeq/L) of surface waters is now occurring in the headwater catchments above GL4 as a result of the increase of inorganic N deposition in wetfall. We illustrate with water quality information from 1994, selected because it is the first year with good NH_4^+ data. ANC concentrations in surface waters draining the 9-ha ARIK catchment were less than 0 μeq/L for three weeks on the rising of the hydrograph during the initiation of snowmelt runoff, with the negative ANC values ranging from -3.8 to -7.0 μeq/L (Figure 8). On the receding limb of the hydrograph ANC values then recovered to a seasonal maximum of 21 μeq/L. At the 42-ha NAV site, ANC concentrations were quite similar to those at the ARIK site, with a

three-week period with values less than 0 μeq/L and an annual maximum value of 21 μeq/L. The ANC values at GL4 were always positive, and ranged from a minimum of 22 μeq/L on the rising limb of the hydrograph to 48 μeq/L on the recession limb of the hydrograph.

The pH values at ARIK and NAV suggest that the loss of buffering capacity is causing a decrease in pH (Figure 8). Minimum pH values at ARIK were 4.8 and at NAV 5.4. At both sites, maximum annual values of pH were less than 6.0. In contrast, at GL4 pH values were never less than 6.2 and increased to circumneutral values of about 7.0 on the receding limb of the hydrograph.

Elevated NO_3^- concentrations are associated with the negative ANC's and low pH's at both the ARIK and NAV sites (Figure 8). At both sites, maximum NO_3^- concentrations are associated with the first fraction of snowmelt runoff and are about 50 μeq/L, or about 5-fold greater than the volume-weighted mean concentrations in the snowpack. At ARIK, concentrations then decrease to an annual minimum of about 5 μeq/L during the growing season before increasing again during the fall. Nitrate concentrations at GL4 track those of the ARIK site quite closely, except that maximum concentrations of NO_3^- at GL4 of 27 μeq/L are about half those of ARIK. Interestingly, minimum concentrations of NO_3^- of 15 μeq/L during the growing season at the NAV site are two to three times the minimum concentrations at either ARIK or GL4.

Nitrification of NH_4^+ released from storage in the seasonal snowpack may partially explain this pattern. Ammonium concentrations in stream waters at ARIK generally ranged from about 5 to 10 μeq/L, with a maximum of about 15 μeq/L (Figure 8). Downstream at the NAV and GL4 sites, NH_4^+ concentrations were always below or near the detection limit of 0.7 μeq/L. Ammonium released from storage in snow at the ARIK site was assimilated by biota or captured on soil exchange sites before reaching the NAV site in stream flow. Nitrification of this NH_4^+ and subsequent transport of NO_3^- to the NAV site may have contributed to the elevated levels of NO_3^- at the NAV site during the growing season. Furthermore, the rapid loss of NH_4^+ from surface waters provides evidence that these high-elevation catchments are not simple flow through systems with respect to the inorganic N stored and released from the seasonal snowpack.

Water Quality: Synoptic Lake Samples

Elevated NO_3^- levels in Green Lakes Valley were associated with a decreasing trend in ANC at GL4 and episodic acidification at the ARIK and NAV sites. The synoptic survey of water quality from lake outflows in 1995 provides information on whether the elevated NO_3^- concentrations we report for the Green Lakes Valley are a site specific problem or are a regional phenomenon in the Colorado Front Range.

Lakes in Wilderness Areas of the Colorado Front Range had greater NO_3^- concentrations than lakes from Wilderness Areas to the west of the Continental Divide. Mean concentrations of NO_3^- from lake outlets in Wilderness Areas of the Front Range were remarkably similar, ranging from 6.3 μeq/L in the Rawah Wilderness Area (n = 19) to 8.1 μeq/L in the Indian Peaks Wilderness Area (n = 16) (Figure 9). In contrast, the mean concentration of NO_3^- from lake outlets in Wilderness Areas to the west of the Front Range was 1.9 μeq/L (n = 19). A one-way analysis of variance shows that NO_3^- concentrations collected on the eastern slope of the Front Range were significantly greater than the western slope at α = 0.05 (p = 0.006). The range of mean NO_3^- concentrations of 6.3 to 8.1 μeq/L in lakes of the Colorado Front Range were quite similar to NO_3^- concentrations at GL4 of 5 to 8 μeq/L during the same time period. The elevated NO_3^- concentrations we report for the Green Lakes Valley occur throughout Wilderness Areas of the Colorado Front Range but not in Wilderness Areas to the west of the Colorado Front Range.

DISCUSSION

Nitrogen Emissions and Deposition

Atmospheric deposition of inorganic N in wetfall to the central Rocky Mountains is relatively modest but greater than background levels. Agriculture, combustion of fossil fuels, and other human activities have altered the global cycle of N substantially, generally increasing both the availability and mobility of N over large regions of the Earth [Vitousek et al., 1997]. Inorganic N deposition in annual wetfall from unpolluted regions of the world generally ranges from 0.1 to 0.7 kg ha^{-1} yr^{-1}, based on extensive measurements of precipitation chemistry in remote areas of the southern hemisphere [Galloway et al., 1982, 1996; Likens et al., 1987; Hedin et al., 1995]. The 2.5 to 3.5 kg ha^{-1} yr^{-1} of inorganic N deposition in wetfall to the Colorado Front Range is about 5-10-fold greater than background amounts. Compared to the northeastern United States, deposition of inorganic N in wetfall to the Rocky Mountains is similar to the 3.7 kg ha^{-1} yr^{-1} at Acadia National Park and about half of the 5.1 kg ha^{-1} yr^{-1} at the Hubbard Brook Experimental Forest as measured over the last ten years by the NADP program [Williams et al., 1998].

Orographic precipitation in the Rocky Mountains compounds the N-loading problem. In the northeastern United States, increases in NO_3^- leaching losses have been associated with high-elevation sites because of higher amounts of N deposition in wetfall [Driscoll et al., 1987]. It is well-documented that many high-elevation ecosystems receive higher doses of nutrients and pollutants than adjacent low-elevation ecosystems in the northeastern United States [Lovett, 1994; Lovett and Kinsman, 1990]. The continental divide in the Colorado Rocky Mountains is generally higher than 4,000 m in elevation, with 52 peaks in Colorado exceeding 4,267 m in elevation. As air masses rise to over 4,000 m in the Rocky Mountains from surrounding lowlands, they cool adiabatically with resulting precipitation as air temperatures decrease below the dew point. Consequently, annual precipitation generally increases with increasing elevation in the Rocky Mountains [Barry, 1973]. In turn, this increase in orographic precipitation with increasing elevation

results in more N deposition in mountain areas compared to surrounding lowlands (Figures 4,5). As in the northeastern US, the highest rates of N deposition in the Rocky Mountains occur at the highest elevations. This orographic effect is much greater than in the northeastern US, since the Rocky Mountains are generally two to three times the height of mountains in the northeast. Consequently, in high-elevation areas of the Rocky Mountains even modest increases in the atmospheric content of anthropogenically-produced N will result in much greater deposition in wetfall of inorganic N compared to the northeastern US.

Dryfall of N as gases, aerosols, and particles provides additional sources of inorganic N in atmospheric deposition. For the Loch Vale watershed in Rocky Mountain National Park, 50 km to the north of Green Lakes Valley, Baron and Campbell [1997] estimate dryfall of N as half of that measured in wetfall. Sievering et al. [1992, 1996] estimate dryfall to Niwot Ridge and Green Lakes Valley as about half to the same as measured N in wetfall. While we recognize dryfall as an important source of inorganic N in atmospheric deposition, we restrict our analyses to inorganic N deposition in wetfall because of the many more and comparable measurements of wetfall. However, we recognize that total inorganic N from atmospheric deposition is potentially 50% to 100% higher than in wetfall alone.

Attributing the inorganic N in atmospheric deposition to specific sources is difficult. During summer months, easterly upslope winds often flow from the metropolitan Denver area and surrounding agricultural areas to the eastern slopes of the Colorado Front Range, transporting urban and agricultural emissions to high elevations [Parrish et al., 1990; Langford et al., 1992; Baron and Denning, 1993; Baron and Campbell, 1997]. During winter months, the predominant westerly winds contribute snowfall from sources to the west of the Colorado Rockies, possibly including point sources such as the Craig and Hayden power plants [Turk et al., 1992]. Using 2,300 back-trajectory analyses, Sievering et al. [1996] suggest that much of the N deposition at Niwot Ridge is from the greater continental area to the west of the Rocky Mountains, particularly southern California. Our emission maps show large amounts of NO_x emissions to the west, east, and south of the central Rocky Mountains. At present we lack the knowledge to identify the

emission sources that are causing the elevated levels of inorganic N in wetfall to the Rocky Mountains. More research on source-receptor relationships on N deposition in the Rocky Mountains is needed.

Water Quality

We believe that aquatic resources in high-elevation catchments of the Rocky Mountains are being negatively affected at current deposition levels of inorganic N in wetfall. The interaction between N deposition and freshwater acidification is complex. Effects of N deposition are generally decoupled from the N deposition because of the large variety of N species found in air, deposition, watersheds, and surface waters, as well as the myriad of pathways through which N can be cycled in terrestrial and aquatic ecosystems [Stoddard, 1994]. Several field experiments have shown that where increased N additions led to increased NO_3^- mobility, the NO_3^- loss led to losses of nutrient cations and increases in soil and water acidity [McNulty and Aber, 1993; Boxman et al., 1995; Emmett et al., 1995]. Furthermore, nitric acid is highly mobile in snowpacks, so that in many areas it is the predominant strong mineral acid released in snowmelt runoff in both the eastern US [Schaefer et al., 1990] and western US [Williams and Melack, 1991b].

The episodic acidification and decrease in ANC of surface waters that we report was predicted in the early 1980's by Kling and Grant [1984]. At that time there were no reports of acidification of surface waters (ANC < 0 μeq/L) in the Rocky Mountains, but there was evidence that precipitation was becoming more acidic [Lewis et al., 1984]. Kling and Grant [1984] predicted that acidification of surface waters in the Rocky Mountains would be detected first at the highest elevations in the Colorado Front Range, because of limited soil extent and flashy hydrographs at these high elevation sites. Our results confirm the prediction of Kling and Grant [1984]. Furthermore, in Caine's [1995] extensive analysis of temporal trends in water quality in the Green Lakes Valley, he reports on earlier records in the Green Lakes valley that were summarized in Caine and Thurman [1990]. At GL4, records of ANC measured in summer from 1969 to 1971 have an arithmetic mean of 81 μeq/L, about the same as that predicted for the year 1980 by the

regression trend at GL4 (Table 1). This analysis suggests that the decline in ANC at GL4 was initiated in the early 1980's. Analysis at the 8-ha Martinelli subcatchment in the Green Lakes Valley (Figure 1) shows that ANC has declined at the rate of -3.2 μeq/L per year for more than 20 years, similar to the -4.2 μeq/L ANC decline at ARIK (Table 1). At the 750-ha Albion sampling site, ANC is decreasing at a lower rate of -1.5 μeq/L. If current levels of inorganic N deposition in wetfall remain constant or increase, episodic acidification of GL4 will begin sometime in the next decade and the higher-elevation ARIK and NAV sites will become chronically acidified.

Much of the research on hydrochemical responses to increasing amounts of inorganic N in wetfall has been conducted in the northeastern US. High-elevation areas in the Colorado Front Range exhibit many of the traits that characterize areas in the northeastern United States that are sensitive to N deposition. Comparison of our results to these research efforts may provide insights as to why high-elevation areas in the central Rockies are being negatively affected by modest amounts of N deposition in wetfall.

Nitrate leaching to surface waters in the northeastern US has been associated with catchments characterized by shallow soils and those sites which have received little human disturbance (which presumably were close to input-output balance prior to receiving enhanced N deposition) [Stoddard and Murdoch, 1991; Kahl et al., 1993]. In general, Pleistocene glaciation has resulted in catchments of the Colorado Front Range having soils that are limited in area and very shallow when present [Kling and Grant, 1984; Caine and Thurman, 1990; Baron, 1992]. Moreover, these high-elevation areas have received little human disturbance compared to the eastern United States and to Europe. The short-growing seasons, location above treeline, and limited soil development result in little agricultural use, no logging, only limited grazing, and little development. Consequently, it appears that these high-elevation ecosystems were close to input-output balance prior to the enhanced N deposition that we report. Furthermore, the increases of anthropogenically-fixed N in the ambient atmosphere of the Colorado Front Range and resulting increases in the deposition of inorganic N in wetfall result in an uncontrolled experiment of N

fertilization at the catchment level similar to controlled experiments in the northeastern United States [Kahl et al., 1993; Williams et al., 1996b]. As in the northeastern US, limited extent of soils combined with little human disturbance results in the high-elevation catchments of the Rocky Mountains having little capacity to assimilate increases in atmospheric deposition of inorganic N.

Additionally, the storage and release of solutes from the seasonal snowpack in the form of an ionic pulse magnifies the aquatic problems caused by pollutants in wetfall. Here we define an ionic pulse as occurring when the initial fraction of snowmelt has ionic concentrations greater than the bulk average for the snowpack [*e.g.* Johannessen and Henriksen, 1978; Colbeck, 1981]. Williams et al. [1996c] have shown that on Niwot Ridge initial concentrations of NO_3^- in snowmelt at the plot scale (1-m^2) may be as high as 20 times those of bulk snowpack concentrations. At the ARIK site, the elevated NO_3^- and H^+ concentrations in surface waters at the onset of snowmelt are consistent with the storage and release of solutes from the seasonal snowpack in the form of an ionic pulse. Storage and release of solutes from the snowpack in the form of an ionic pulse at the 9-ha ARIK site magnifies the concentration of pollutants in wetfall about 5-fold.

Ammonium released from storage in the seasonal snowpack may also contribute to the observed acidification of surface waters. The presence of NH_4^+ in surface waters at the ARIK site suggests that there was little modification of the chemical content of snowmelt before contributing to stream flow at that site. Additionally, reactive silicate (Si) in surface waters at ARIK was always at or below detection limits (data not shown), indicating that the ARIK site acts to some extent like a snow lysimeter 9-ha in area. However, the lack of NH_4^+ in surface waters at the 42-ha NAV site directly downstream from ARIK shows that the NH_4^+ in stream waters was rapidly immobilized by some combination of geochemical reactions and biological assimilation. Biological assimilation of NH_4^+ results in the production of 2 moles of H^+ for every mole of NH_4^+ assimilated [Reuss and Johnson, 1986]. The neutralization of NH_4^+ deposited in wetfall is thus a likely contributor to acidification of surface waters.

The elevated NO_3^- concentrations that we report for lakes throughout the Colorado Front Range indicate that the changes in surface water quality in the Green Lakes Valley are part of a regional phenomenon. Comparison of NO_3^- concentrations from our synoptic lakes survey to those collected as part of the southern Rocky Mountain section of the Western Lakes Survey conducted in 1985 by the EPA provides additional evidence that elevated NO_3^- concentrations are a regional problem throughout the Colorado Front Range that is not yet occurring in other areas of the Rocky Mountains. Our synoptic survey differs from the Western Lakes Survey (WLS) in two important ways: 1) The WLS was conducted in September and our survey was conducted in late July and early August, and 2) lakes sampled as part of the WLS were selected using a rigorous statistical design and our lakes were determined in part by logistical constraints. Nonetheless, the comparison provides additional insight into the regional nature of elevated NO_3^- concentrations in surface waters of the Rocky Mountains.

There were 104 lakes sampled by the WLS outside of the Colorado Front Range and we sampled 91 lakes within the Colorado Front Range (Figure 10). About 58% of the WLS lakes had NO_3^- concentrations less than 1 μeq/L compared to only 23% of the lakes in our synoptic survey. Clearly, most of the catchments sampled by the WLS were N-limited with little leakage of the NO_3^- to surface waters. In contrast, none of the WLS lakes outside of the Colorado Front Range had NO_3^- concentrations greater than 10 μeq/L, while 30% of our lakes had NO_3^- concentrations greater than 10 μeq/L. Catchments in the Colorado Front Range leak more NO_3^- to surface waters than catchments in other areas of the Colorado Rocky Mountains.

These results suggest that N leakage from terrestrial environments to aquatic systems is occurring throughout the Colorado Front Range at current loading of inorganic N in wetfall. Most other high-elevation areas in the central Rockies are not leaking NO_3^- at this time. However, these areas are in danger of becoming N-saturated in the near future if present levels of N deposition in wetfall continue or increase.

Biological Effects

Degradation of terrestrial and aquatic resources may now be occurring as a direct result of the increased atmospheric deposition of inorganic N in wetfall. Increases in N availability are leading to changes in plant species composition as a result of competitive displacement [Bowman and Steltzer, 1998]. At Niwot Ridge, over the last 40 years there have been statistically significant increases in the vegetative cover of several N-enrichment indicator species [Korb, 1997]. Furthermore, Bowman and Steltzer [1998] have shown that the biotic response to increased N deposition includes a positive feedback mechanism that may further contribute to watershed-level N saturation. In moist meadow communities of alpine tundra on Niwot Ridge, increasing N deposition results in the replacement of the dominant *Acomastylis rossii* by *Deschampsia caespitosa* and an eight-fold increase in net N mineralization and nitrification rates, which in turn will increase groundwater loss of NO_3^- to aquatic ecosystems [Bowman and Steltzer, 1998].

There is some evidence to suggest that high-elevation forests may be at risk to winter damage from increasing N deposition in the Colorado Front Range. In the northeastern United States, high-elevation red spruce show high levels of mortality because of reduced cold tolerance caused by increased amounts of atmospheric pollutants [Craig and Friedland, 1991; Eagar and Adams, 1992]. While increased frequency of damage to red spruce foliage has been notable at high elevations in the northeastern United States, only a very mild form of winter damage has been observed in the South [Anderson et al., 1991], probably because extreme low temperatures occur less frequently at high-elevation areas in the South [NAPAP, 1998]. However, there is uncertainty about the relative importance of sulfur, nitrogen, and acidity in causing this decline in cold tolerance.

In the Colorado Front Range, higher N:P ratios have been reported along an elevational gradient in foliage of bristlecone pine [Williams et al., 1996b]. Similar changes in the N:P ratio of foliage have been reported for Norway spruce in Sweden and other forested sites as a result of elevated N deposition in wetfall [Fenn et al., 1998]. Furthermore, comparisons of forest ecosystems in the Loch Vale catchment of Rocky Mountain National Park with Fraser Experimental

Forest in western Colorado (N deposition of 1-2 kg ha^{-1} yr^{-1}) show that Front Range forests have lower C:N ratios in soils and foliage. Although the forests were matched as closely as possible (500-700 yr spruce-fir, 3100 to 3200 m elevation range), soil C:N ratios of 34.5 in Fraser were about 25% greater than the 24.3 in Loch Vale, with C:N ratios in foliage about 8 to 10 units lower at Loch Vale compared to Fraser [Baron et al., 2000 (in press)]. In general, the response of forested ecosystems to increases in N deposition are not linear and would therefore not be captured in simple dose-response functions [Aber et al., 1998]. The higher N:P ratios and lower C:N ratios in forested ecosystems of the Colorado Front Range, combined with the harsh winter climate, suggest that forested ecosystems in the Colorado Front Range may now be at risk to winter damage.

The acidification of surface waters and resulting decrease in pH can cause changes in the aquatic resources of high-elevation catchments. Zooplankton species, such as the dominant *Daphnia rosea*, begin decreasing below pH 5.5 to 5.8 and virtually disappear below pH 5.0 [Barmuta et al., 1990]. In turn, decreases in the population of *Daphnia rosea* result in increases of more acid-tolerant species such as *Bosmina* spp, resulting in a restructuring of the natural zooplankton assemblages when pH decreases to about 5.5. Among the benthic invertebrates found in western streams, the mayfly larva (*Baetis* spp.) is very sensitive to acidic episodes, with populations decreasing rapidly once pH amounts drop below 5.5 [Kratz et al., 1994]. These species are important as food items for native fish in high-elevation aquatic systems. Native fish species, such as cutthroat trout and rainbow trout, have sensitivity to acidic waters depending on the life stage exposed to acidic episodes. In general, fish population viability is expected to be reduced below pH 6 [Baker et al., 1990]. In the eastern United States, streams with low ANC in Shenandoah National Park (Virginia) showed fish populations with decreased species richness, population density, condition factor, age distribution, and size compared to streams with higher ANC [Bulger et al., 1995; Dennis et al., 1995; MacAvoy and Bulger, 1995]. Furthermore, a study of 13 streams in the Adirondack and Catskill Mountains in New York and the northern Appalachian Plateau in Pennsylvania showed long-term adverse effects on fish populations from episodic

reductions in ANC similar to those we report for the Green Lakes Valley [Wigington et al., 1996]. Fish populations in high-elevation catchments of the Colorado Front Range are at risk of reduced viability as a result of increasing amounts of inorganic N in wetfall.

Two amphibian studies conducted in the Rockies point to the direct effects on salamander eggs of acidic episodes, and possible community level responses of competing amphibian species. Harte and Hoffman [1989] exposed the eggs of tiger salamanders (*Ambystoma tigrinum*) to experimental increases in episodic acidification and determined that they had an LD-50 pH of 5.6, which is within the range of snowmelt pH's encountered in the Rocky Mountains and higher than pH's we report during snow melt runoff. In an experimental study of coexisting populations of tiger salamanders and chorus frogs (*Pseudarcris triseriata*), Kiesecker [1996] reports the possibility that changes in development rates in these larvae can be affected by depressed pHs in pond water, leading to changes in predation success.

Adverse biological effects may be occurring at present levels of N deposition in wetfall and the resulting reductions in ANC. Furthermore, chemical effects in surface waters due to changes in atmospheric deposition can exhibit lag times of many years. Lags in measurable effects on aquatic biota can be even longer [NAPAP, 1998]. If current amounts of N deposition in wetfall to the Colorado Front Range continue or increase it is possible that negative biological effects will occur and intensify.

Critical Loads

Water quality in high-elevation catchments of the Colorado Front Range is being degraded at present levels of N deposition in wetfall. Protected areas such as National Parks and Wilderness Areas with elevated amounts of N deposition are susceptible to N leakage because of stand maturity where forested, accumulation of N in soil, and particularly in high-elevation sites, low N retention capacity of soils and vegetation. All of these factors suggest that reductions of N emissions are needed to protect these environments [Fenn et al., 1998].

Our challenge, from a scientific perspective, is to design research and monitoring efforts that will provide a credible foundation for regulatory action in a timely fashion. The National Park Service (NPS) and other Class 1 federal land managers are guided by a fundamental, legislatively-mandated objective: to protect Class 1 resources from the adverse effects of air pollution so as to leave them unimpaired for future generations [Bunyak, 1993]. The land manager is congressionally directed to err on the side of protection. However, given the political and economic ramifications of policy decisions, land managers are acutely aware of the need to provide a rational, scientific basis for those decisions [Shaver et al., 1994].

At present in the western United States, no "critical loads" have been established for N deposition in wetfall. However, there has been discussion within the EPA and movement in some states to set "deposition standards" or "secondary standards" that would protect sensitive resources, such as low-alkalinity surface waters and already acidic soils. Finally, there is a lack of approved methods for quantifying what effect incremental degrees of pollution would have on terrestrial, aquatic, and visibility resources. Demonstrating the "adverse" effect of a small change in emissions, when current pollutant loadings are already causing harm to air-quality related values (AQRVs) is no small task, scientifically or politically.

Researchers have proposed a six-step approach for setting critical loads in the United States: (1) selection of ecosystem components, indicators, and characterization of the resource; (2) definition of functional subregions; (3) characterization of deposition within each of the subregions; (4) definition of an assessment endpoint; (5) selection and application of models; and (6) mapping of projected ecosystem responses [Holdren et al., 1993; Hunsaker et al., 1993]. We believe that the empirical information that we have presented are sufficient to propose critical loads for inorganic N deposition in wetfall to the central Rocky Mountains, because of the wealth of site-specific and synoptic data for high-elevation sites in this region.

The ecosystem components and indicators that we believe are important for this evaluation are concentrations of inorganic N in surface waters and the pH and ANC of those surface waters. The region of interest is high-elevation catchments in the central Rocky Mountains, with special

emphasis on the subregion of the Colorado Front Range. Deposition amounts of inorganic N in wetfall to this region and subregion range from 2.5 to 3.5 kg ha^{-1} yr^{-1}. This annual deposition of inorganic N in wetfall is sufficient to cause increased concentrations of NO_3^- in many surface waters in the region. In turn, the elevated concentrations of NO_3^- in surface waters are causing episodic acidification (ANC < 0) and chronic and sustained decreases in ANC in the Green Lakes Valley on an annual basis. The lack of long-term data from other sites in high-elevation areas makes it impossible to determine if episodic acidification and/or decreases in ANC are occurring throughout the Colorado Front Range.

Balancing the federal mandate of land managers to protect natural resources in Class 1 areas with political and economic realities, we take a conservative approach and suggest that critical loads of inorganic N deposition in wetfall to Class 1 areas in the central Rocky Mountains be set at 4 kg ha^{-1} yr^{-1}. While episodic acidification of headwater catchments is now occurring in Green Lakes Valley at present deposition levels of inorganic N in wetfall of about 3.5 kg ha^{-1} yr^{-1}, we do not know if this amount of inorganic N deposition will cause episodic acidification elsewhere in the Colorado Front Range because of site specific differences (bedrock type, soil type and development, slope angle, aspect, etc). Conversely, for these same reasons it is quite possible that streams and lakes in some areas of the Colorado Front Range may experience episodic acidification at lower levels of inorganic N deposition in wetfall than is presently occurring in Green Lakes Valley. Target loads may be set by federal land managers at lower levels of N deposition to allow a margin of safety to protect these very sensitive resources. While an approach based on critical loads may not be appropriate conceptually for some of the consequences of N deposition-it is not clear that there are thresholds for all of these effects-critical loads represent a useful tool for managing N deposition [Vitousek et al., 1997]. The critical load that we propose is comparable to the 3-8 kg ha^{-1} yr^{-1} critical load of N deposition proposed for forested catchments in granitic basins of Europe, based on results from ecosystem-scale experiments (NITREX) [Reynolds et al., 1998].

Modeling of critical loads would address some of the limitations caused by our empirical approach. Furthermore, we recognize that in many other mountain regions of North America that this kind of long-term, intensive data on watersheds and surface waters will not be available. In such cases, data collected at intensive sites can be incorporated into models of watershed responses to increased amounts of inorganic N in wetfall. These models can then be used to simulate and spatially distribute the effects of increases in deposition of inorganic N at sites with more limited data availability. Critical loads for inorganic N deposition can also be generated in this way, with the possibility of using model simulations to perform regional assessments. Such an approach is being tested in National Parks and Wilderness Areas of the western US using MAGIC-WAND (Model of Acidification of Groundwater in Catchments, with Aggregated Nitrogen Dynamics) [Ferrier et al., 1995].

Mitigation of adverse ecosystem effects in Class 1 areas is not an acceptable alternative to emission controls. Federal and state agencies have been mandated to adopt management prescriptions known as best management practices (BMPs) to regulate land uses to achieve water quality standards [Whitman 1989]. However, the concept of BMPs is antithetical to management of Class 1 areas. Congress, under the Clean Air Act Amendments, has stated that there is no amount of resource damage from atmospheric pollution that is acceptable in Class 1 areas.

Degradation of water quality and biological resources in the alpine High Tatra Mountains of central Europe from atmospheric deposition of pollutants provides an example of what may happen if critical loads for N deposition in the Rocky Mountains are not enacted. The High Tatra Mountains are located within 200 km of the so-called Black Triangle of the German-Polish-Czech border area. Elevated levels of N deposition resulted in N saturation of high-elevation catchments with granitic basins and chronic acidification of surface waters in the High Tatra Mountains [Kopacek et al., 1995]. Sulfur and N emissions have been reduced by 30% to 40% in Central Europe since 1989 due to the political and economic changes in the postcommunist countries [Kopacek et al., 1998]. At Vysne Wahlenbergovo Lake in the High Tatra Mountains, the reduction in N emissions has resulted in NO_3^- concentrations decreasing from about

50 μeq/L to 30 μeq/L and a sharp increase in the pH of lake water [Kopacek et al., 1998]. The rapid decrease in NO_3^- concentrations suggests that N saturation of these watersheds was related to elevated levels of atmospheric deposition of N that exceeded the assimilation capacity of the ecosystem. Unfortunately, the improvement of water quality, particularly pH, has not yet been sufficient to bring about significant change in the biota and the return of planktonic crustacea that have become extinct [Kopacek et al., 1998]. We suggest that critical loads for N deposition be enacted for Class 1 areas in the Rocky Mountains before large-scale resource damage occurs.

SUMMARY

High-elevation catchments in the Rocky Mountains and other mid-latitude regions of the world are sensitive to atmospheric deposition of pollutants because of: (i) increased deposition loading because of increased orographic precipitation amount; (ii) increasing acidity of precipitation with elevation because of washout of neutralizing base cations; and (iii) release of pollutants from the snowpack in the form of an ionic pulse, which magnifies the concentration of pollutants stored in the seasonal snowpack. These processes, exacerbated by large point and non-point sources of inorganic N emissions in and near the Rocky Mountains, have resulted in episodic acidification and NO_3^- leakage in the Green Lakes Valley of the Colorado Front Range at present deposition levels of inorganic N in wetfall.

Federal land managers, in collaboration with the EPA and states, will continue to manage parks and wilderness areas using more established air pollution control policy tools, such as Prevention of Significant Deterioration (PSDs) and National Ambient Air Quality Standards (NAAQS). However, because of the regional nature of the pollutants affecting protected areas, such as atmospheric deposition of inorganic N, the future of air management will be to progress toward regional control of sources and setting of critical loads to prevent damage to sensitive resources.

Here we propose critical loads for N deposition in wetfall to the Colorado Front Range of 4 kg ha^{-1} yr^{-1}, as a regional threshold that cannot be exceeded if we are to protect high-elevation

ecosystems. We note that this critical load range for inorganic N deposition in wetfall will be refined as more data on sensitive ecosystems are collected. We further note that Federal Land Managers, who have "the affirmative responsibility to err on the side of protection", may well set a more restrictive target load to protect the most sensitive waters and soils from changes caused by inorganic N deposition in wetfall.

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Table 1. Alkalinity decreases in surface waters of the Green Lakes Valley, 1984-1995.

Site	N	Slope	Alkalinity Decrease (μeq $L^{-1}yr^{-1}$)	Significant at $p < 0.05$
Arikaree	130	-0.0115	-4.2	Yes
Martinelli	160	-0.0087	-3.2	Yes
GL4	260	-0.0069	-2.5	Yes
Albion	262	-0.0041	-1.5	No

FIGURES

Figure 1. Location map showing the intermountain West, Colorado Front Range, Wilderness Areas in and near the Colorado Front Range, and a topographic map of the Green Lakes Valley and Niwot Ridge. ARIK is the 9-ha Arikaree sampling site; NAV is the 42-ha Navajo sampling site; and Green Lake 4 (GL4) was sampled at its outlet. An NADP wet deposition site is located at the Saddle on Niwot Ridge, which forms the northern boundary of Green Lakes Valley.

Figure 2. Location of 58 point sources that produce more than 1,000 tons per year of NO_x in and near the Rocky Mountains in the US. The 2,800 m contour of the Rocky Mountains is the thicker black line. Class 1 National Park areas are shown in grey.

Figure 3. County NO_x emissions for the intermountain West region, 1990. Urban centers with populations greater than 50,000 are shown as thin black lines. The greatest emissions rates (greater than 5000 tons/year) are generally located in the vicinity of the population centers, where the NO_x sources are largely vehicular.

Figure 4. A map of the average annual loading of inorganic N in wet deposition from 1992-1996 for all NADP sites in the intermountain West clearly illustrates that montane areas receive more loading than do low elevation areas (Niwot Ridge NADP data corrected as described in Williams et al.[1998]). All NADP sites in Arizona, New Mexico, and Utah (except one) are less than 2,500 m in elevation and received less than 2.5 kg ha^{-1} yr^{-1} of inorganic N deposition for this time period. In contrast, montane sites above 2,500 m in Colorado and Wyoming received more han 2.5 kg ha^{-1} yr^{-1} of inorganic N for the same time period.

Figure 5. Changes in precipitation amount, annual volume-weighted mean concentrations of NO_3^- (μeq/L), and NO_3^- loading (kg ha^{-1}), for three NADP sites along an elevational gradient in 1994: Pawnee site at 1,641 m in the Great Plains located just east of Rocky Mountain National Park; Beaver Meadows site at 2,490 m on the eastern edge of Rocky Mountain National Park; and the Loch Vale site at 3,159 m near the continental divide in Rocky Mountain National Park. Precipitation amount increases with elevation, concentrations generally decrease with elevation, while chemical loading from wet deposition increases with elevation.

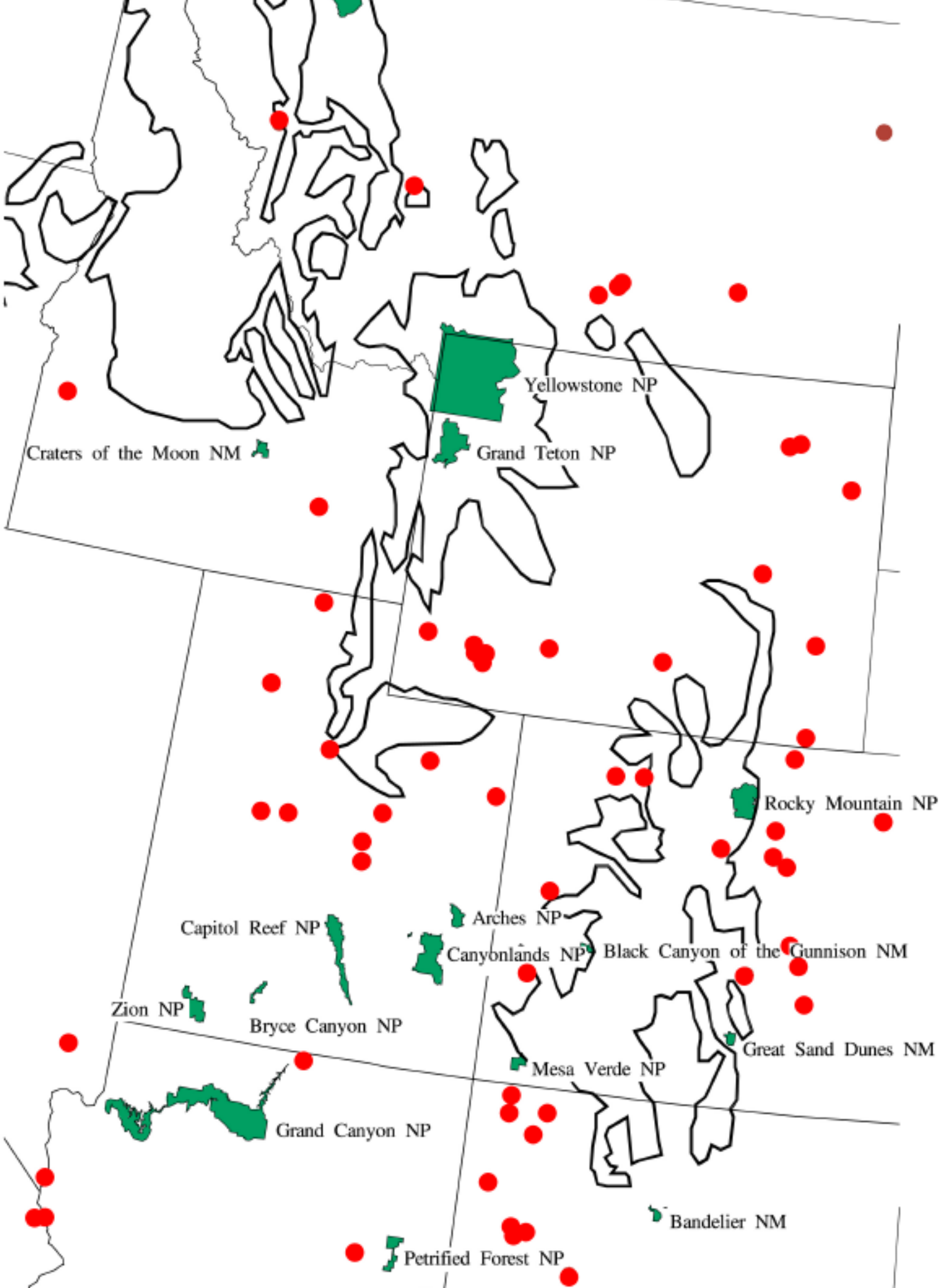
Figure 6. Time series of: a) annual inorganic N deposition in wetfall (kg ha^{-1} yr^{-1}) as measured at the Niwot Ridge NADP site; b) annual minimum concentrations of NO_3^- from the outlet of Green Lake 4; and c) NO_3^- concentrations from the outlet of Green Lake 4.

Figure 7. Time series of ANC concentrations at the outlet of GL4. A simple linear regression of ANC versus time shows that alkalinity in GL4 has been decreasing at the rate of 2.5 μeq/L per year ($r^2 = 0.24$, slope = -0.0069, $p < 0.05$).

Figure 8. A time series of ANC, pH, NO_3^-, and NH_4^+ concentrations in surface waters from the 9-ha ARIK site, the 42-ha NAV site, and the 220-ha GL4 site in Green Lakes Valley, 1994. Acid neutralizing capacity values were below 0 μeq/L for parts of 1994, showing that episodic acidification has begun in the Green Lakes Valley.

Figure 9. Mean concentrations of NO_3^- from high-elevation lakes in wilderness areas of the Colorado Front Range (Indian Peaks (IN), n = 16; Mount Evans (ME), n = 7; Rawah (RW), n = 19; and Sangre de Cristo (SC), n = 50) were significantly greater ($p = 0.006$) than mean NO_3^- concentrations from lakes in wilderness areas to the west (WS) of the Colorado Front Range (WS, n = 19).

Figure 10. Comparison of NO_3^- concentrations from all lakes sampled (n = 104) as part of the southern Rocky Mountains section of the Western Lakes Survey in 1985 (except those lakes in the Colorado Front Range) to all lakes (n = 91) in the Colorado Front Range sampled as part of our synoptic survey in 1995.


Yellowstone NP
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Rocky Mountain NP
Capitol Reef NP
Arches NP
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Mesa Verde NP
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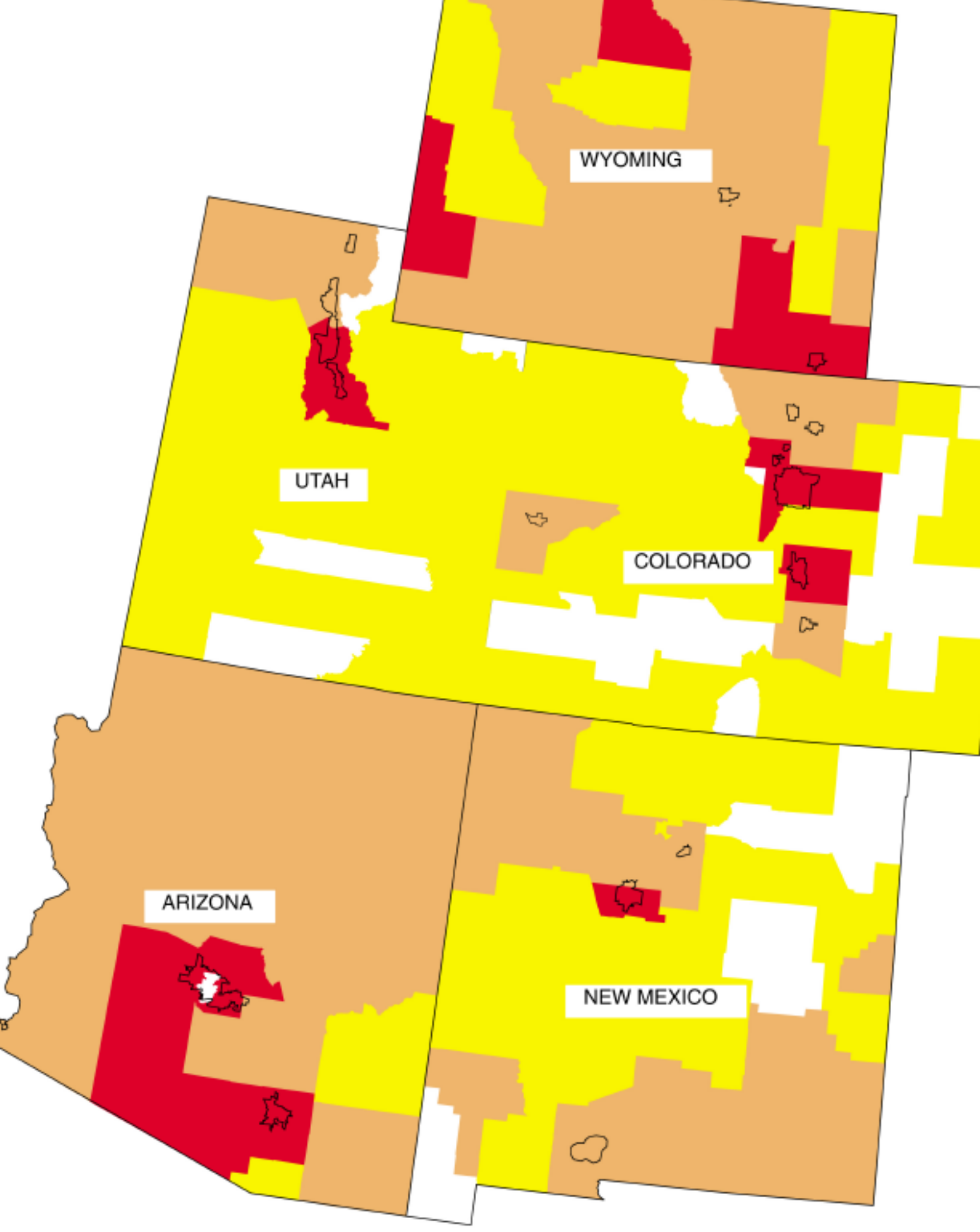
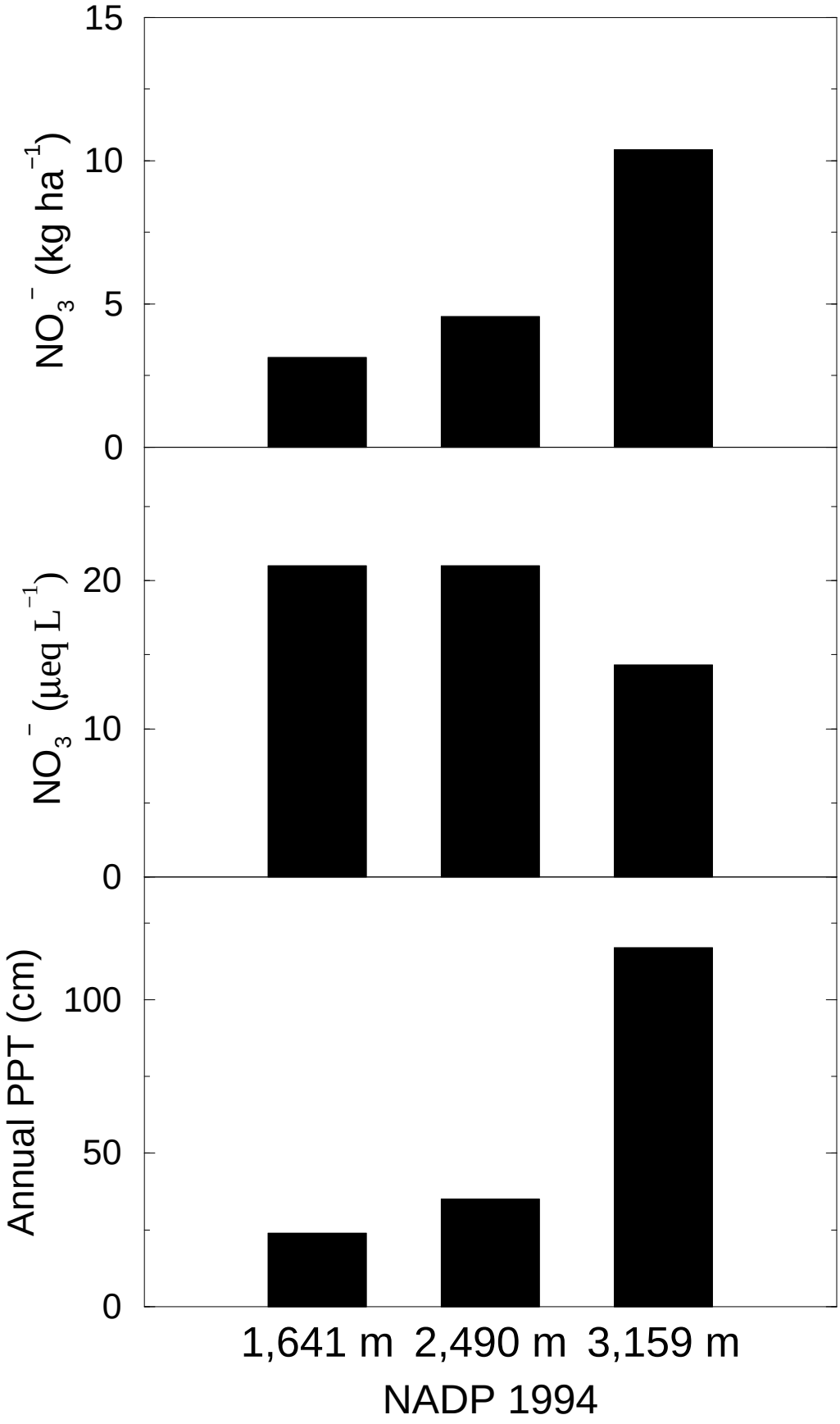
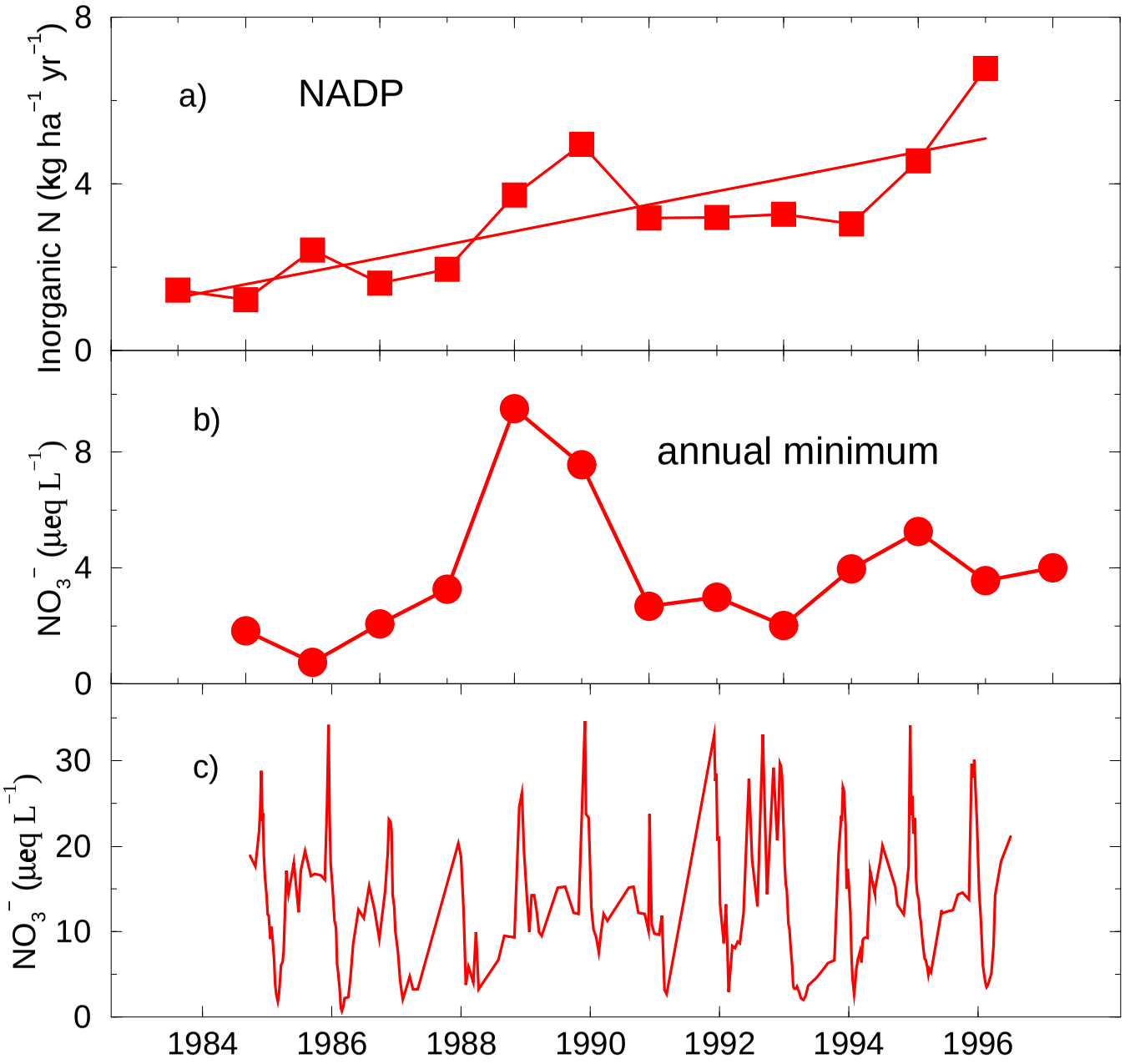
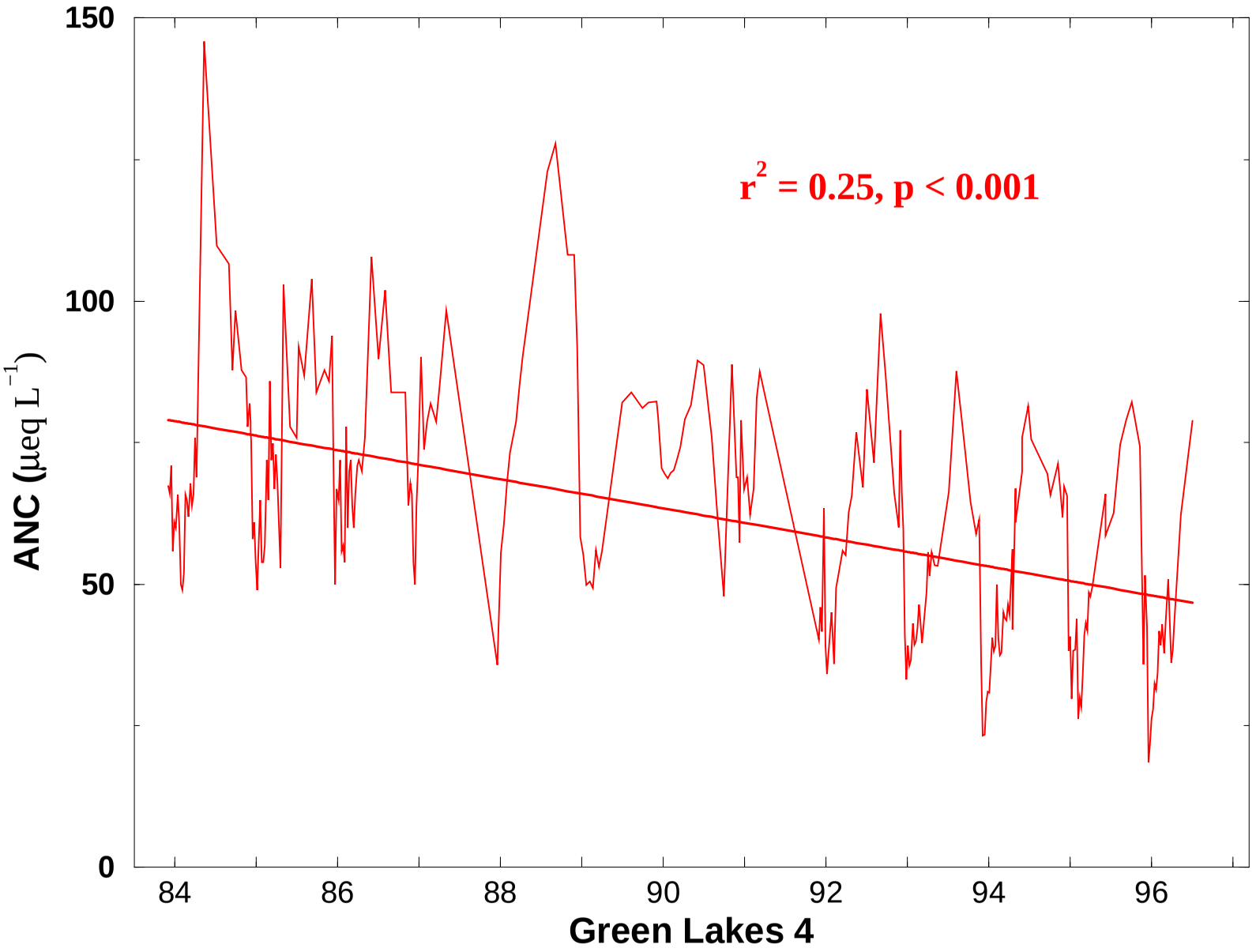
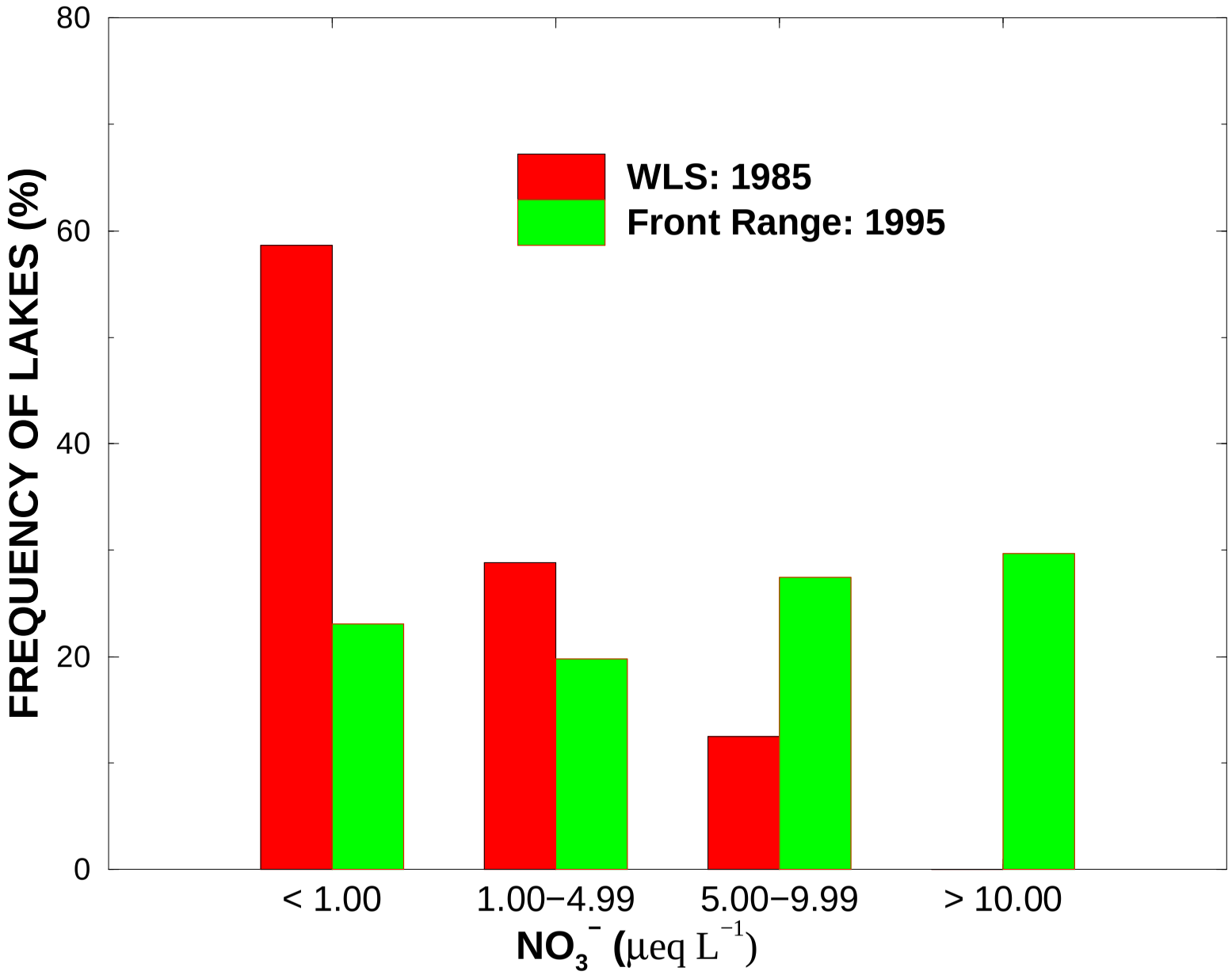
Bandelier NM
Petrified Forest NP


WYOMING
UTAH
COLORADO
ARIZONA
NEW MEXICO


15
10
5
0
NO_3^- (kg ha^{-1})
20
10
0
NO_3^- (µeq L^{-1})
100
50
0
Annual PPT (cm)
1,641 m
2,490 m
3,159 m
NADP 1994


a)
NADP
Inorganic N (kg ha^{-1} yr^{-1})
0
4
8
b)
annual minimum
NO_3^- (μeq L^{-1})
0
4
8
c)
NO_3^- (μeq L^{-1})
0
10
20
30
1984
1986
1988
1990
1992
1994
1996


150
100
50
0
ANC (μeq L^{-1})
r^2 = 0.25, p < 0.001
84
86
88
90
92
94
96
Green Lakes 4


80
60
40
20
0
FREQUENCY OF LAKES (%)
WLS: 1985
Front Range: 1995
< 1.00
1.00−4.99
5.00−9.99
> 10.00
NO3− (μeq L−1)