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Atmospheric deposition inputs and effects on lichen chemistry and indicator species in the Columbia River Gorge, USA

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Nitrogen, sulfur and acidic deposition threaten natural and cultural resources in the Columbia River Gorge National Scenic Area.

Abstract

Topographic and meteorological conditions make the Columbia River Gorge (CRG) an 'exhaust pipe' for air pollutants generated by the Portland-Vancouver metropolis and Columbia Basin. We sampled fog, bulk precipitation, throughfall, airborne particulates, lichen thalli, and nitrophytic lichen distribution. Throughfall N and S deposition were high, 11.5–25.4 and 3.4–6.7 kg ha^{-1} over 4.5 months at all 9 and 4/9 sites, respectively. Deposition and lichen thallus N were highest at eastern- and western-most sites, implicating both agricultural and urban sources. Fog and precipitation pH were frequently as low as 3.7–5.0. Peak NO_x, NH_3, and SO_2 concentrations in the eastern CRG were low, suggesting enhanced N and S inputs were largely from particulate deposition. Lichens indicating nitrogen-enriched environments were abundant and lichen N and S concentrations were 2× higher in the CRG than surrounding national forests. The atmospheric deposition levels detected likely threaten Gorge ecosystems and cultural resources.
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Keywords: Ammonium; Nitrate; Sulfate; Pacific Northwest; Passive sampler; IMPROVE; NADP

1. Introduction

In recent years, nitrogen deposition effects have been documented for various ecosystems of western North America (Fenn et al., 1998, 2003), including impacts on lichen communities along the Columbia River Gorge, located along a portion of the Oregon/Washington border. The Columbia River Gorge National Scenic Area (Fig. 1), highly valued for its scenic, cultural, and natural resources, has become a conduit of air pollution from emissions sources to the east and west (Fig. 2). To the east, the Columbia Basin and counties along the Columbia River, experience relatively frequent air stagnation episodes in winter. During these days- to weeks-long events, emissions are trapped by geographic barriers and capped by regional temperature inversions, preventing vertical mixing or transport. The cold, dense, humid air settles onto the valley floor to the east, then slowly follows the river channel westward through the Scenic Area. This cold air mass drainage is often accompanied by low clouds and fog that entrain air pollutants that are trapped in the Columbia Basin. In summer, the flow reverses as pressure off the coast pushes the air eastward, funneling pollutants through the steep-walled Columbia Gorge from the adjacent Portland-Vancouver-Longview metropolitan areas.

An analysis of 2001 emissions (US EPA, 2005) from counties most likely to contribute to pollution in the Scenic Area (Fig. 2) shows that Scenic Area counties have lower NO_x, NH_3 and SO_2 emissions than bordering counties to the west and east. NO_x emissions are highest from the urban,

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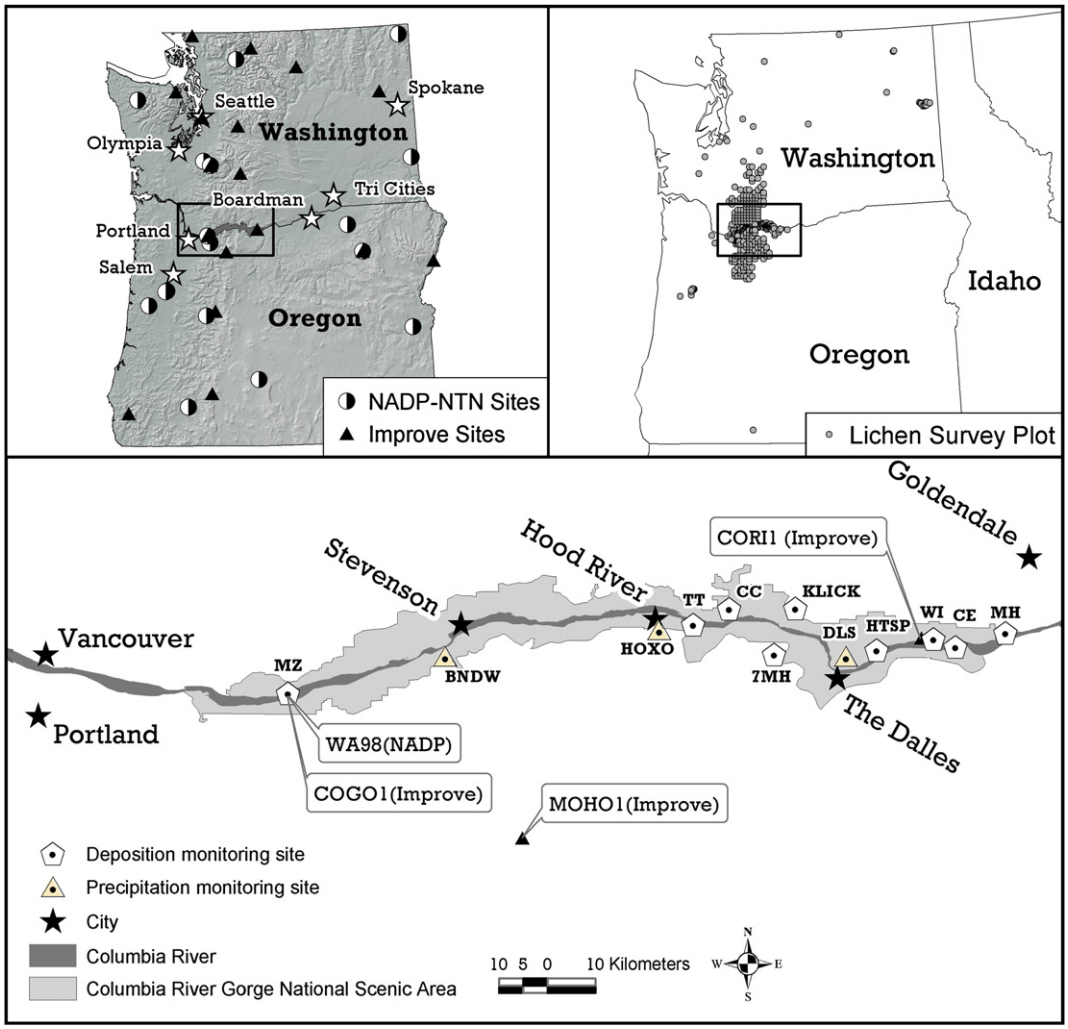

Fig. 1. Maps of air quality monitoring sites and geographic features in the Columbia River Gorge in Oregon and Washington. Monitoring site name abbreviations are given in Table 1 and abbreviations for weather stations are given in the footnote to Table 2. At the Twin Tunnels (TT) and at the Catherine Creek (CC) sites, deposition samples were collected at a relatively low and high elevation location (see Table 1 for more detail).

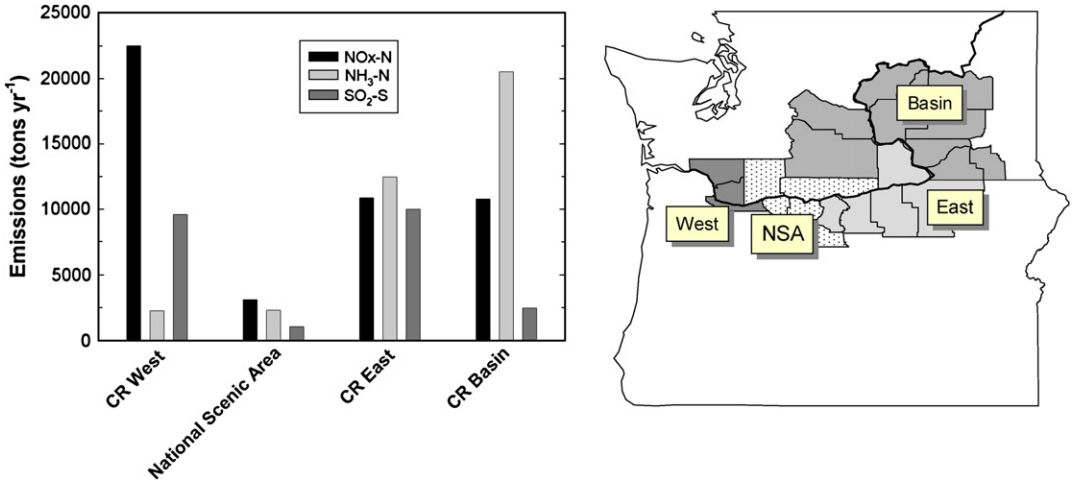

Fig. 2. Total 2001 emissions (US EPA, 2005) of nitrogen and sulfur from criteria pollutants within and upwind of the Columbia River Gorge National Scenic Area (NSA). Emissions values are expressed as N or S. NO_x emissions were reported as NO_2. Pollutants are funneled through the Scenic Area in both directions. The four county groupings shown on the map correspond to the four groups of emissions data in the bar graph (Columbia River West, NSA, CR East and CR Basin).

industrialized western counties, NH_3 emissions are highest from the predominantly agricultural eastern and Columbia Basin counties, and total SO_2 emissions are similar east and west of the Scenic Area. The largest single NO_x and SO_2 source in the 'West' county group of Fig. 2 are commercial marine vessels (18 301 and 3511 tons per year). The largest single NO_x and SO_2 source in the 'East' county group is the Boardman coal-fired power plant located ca. 100 km east of the Scenic Area (10 768 and 17 821 tons per year). Highway vehicles (39%), off-highway vehicles (36%; including agriculture & construction equipment, boats and trains,) and industry (22%) contribute 97% of total NO_x emissions across all counties, and 78% of total SO_2 emissions are from industrial sources. Cattle and dairy production (52%), application of crop fertilizers (29%), industry (10%), and mobile sources (5%) account for 96% of total NH_3 emissions. Total SO_2, NO_x and NH_3 emissions from the counties shown in Fig. 2 were 46 000, 155 000, and 45 500 tons, respectively (US EPA, 2005).

Protection of cultural resources from the effects of air pollution, particularly historic petroglyphs and pictographs of the Columbia Plateau Indians, is a major concern. These historic sites are frequently immersed in polluted, potentially acidic, air masses in the Scenic Area. Pollution exposures in these sites have not been well characterized, total deposition inputs have not been measured, and the effects of air pollution on cultural resources in the Scenic Area are largely unknown. A strong nitrogen (N) air pollution signature in the Scenic Area has been detected from lichen element concentrations and community composition (Fenn et al., 2003; Geiser and Neitlich, in press), raising concerns about effects of atmospheric deposition to other natural resources in the Columbia River Gorge (CRG). Symptoms of N saturation or N excess, as a result of chronic atmospheric N deposition, have been reported in some areas of California and Colorado (Fenn et al., 1998, 2003), but this has not been investigated in the CRG.

The purpose of this study was to evaluate air quality status in the Columbia River National Scenic Area for N and sulfur (S) containing pollutants by integrating results from passive, active, and biological sampling techniques. We hypothesized that spatial patterns of N and S deposition would correspond to patterns of lichen chemistry and indicator species responses. To test this hypothesis, we determined N and S levels in precipitation, canopy throughfall, fog water, ambient air, and lichen thalli, and we also measured deposition acidity. We surveyed lichen species indicative of eutrophic environments to detect ecological responses. Finally, we assessed, from a regional perspective, Scenic Area N and S pollutants using wet deposition data from the National Atmospheric Deposition Program, National Trends Network (NADP/NTN) and aerosol chemistry data from the Interagency Monitoring of Visual Protected Environments (IMPROVE) visibility network.

2. Materials and methods

2.1. Deposition and pollutant monitoring

Atmospheric deposition monitoring was carried out at eleven sites across the CRG (Fig. 1). Site names, abbreviations, elevation and types of samples collected at each site are given in Table 1. The most westerly site was Mt. Zion (MZ), a NADP monitoring site near Portland. The other ten sites were in the eastern half of the Scenic Area (Fig. 1). This study emphasized the eastern side of the Scenic Area because this is where a large number of historic petroglyphs and pictographs, possibly at risk from acidic deposition, are located and because during the winter wet season the primary source region is to the east of the Scenic Area, because of prevailing easterly winds. It was hypothesized that the Columbia Basin, east of the Scenic Area, is an important pollutant source region in winter as suggested by IMPROVE data and previous lichen monitoring (Geiser and Neitlich, in press). Precipitation decreases dramatically from west to east across the Scenic Area. From Bonneville Dam, near MZ to The Dalles (Fig. 1), annual precipitation in the 2003/2004 water year (October 1, 2003 to September 30, 2004) decreased from 176 cm to 34 cm (Table 2). Although the Scenic Area refers to a political boundary and the Columbia River Gorge to a geographic area, the terms are herein used synonymously.

Bulk precipitation and fog water were collected in open areas while bulk throughfall was collected under ponderosa pine (*Pinus ponderosa* Dougl. ex P. & C. Laws) canopies. Bulk deposition, throughfall and fog samples were collected at eleven, nine, and seven sites, respectively (Fig. 1 and Table 1). Sampling occurred from late October 2003 through early March 2004, a total of 19 weeks. Three replicate bulk deposition collectors were installed at each site. Ten throughfall collectors were installed under 5–7 trees per site, except at Celilo (CE), where four throughfall collectors were placed under the single large pine tree.

Table 1
Deposition and lichen elemental analysis collection sites within the Columbia River Gorge

Site Name	Site abbreviation and type of samples collected[a]	Elevation (m)	Coordinates
Mt. Zion	MZ (bp, f, le, NADP, IMPROVE)	225	N 45° 34′ 9.84″ W 122° 12′ 41.04″
Twin Tunnels High	TTH (bp, tf, le)	164	N 45° 41′ 32.77″ W 121° 27′ 01.97″
Twin Tunnels Low	TTL (bp, tf, le, ls)	115	N 45° 41′ 25.94″ W 121° 26′ 48.29″
Catherine Creek High	CCH (bp, tf, le, ls)	385	N 45° 43′ 20.66″ W 121° 22′ 58.98″
Catherine Creek Low	CCL (bp, tf, le, ls)	125	N 45° 42′ 45.28″ W 121° 21′ 57.34″
Seven Mile Hill	7MH (bp, f, tf, le)	520	N 45° 38′ 13.20″ W 121° 17′ 55.72″
Klickitat	KLICK (bp, tf, le, ls)	207	N 45° 43′ 23.76″ W 121° 15′ 32.62″
Horsethief State Park	HTSP (bp, f, tf, wd, le, ls)	33	N 45° 38′ 41.42″ W 121° 06′ 26.87″
Wishram	WI (bp, f, le, ls, IMPROVE)	232	N 45° 39′ 56.98″ W 121° 00′ 2.59″
Celilo	CE (bp, f, tf, ls)	19	N 45° 39′ 03.89″ W 120° 57′ 34.73″
Maryhill	MH (bp, f, tf, ls)	216	N 45° 40′ 37.36″ W 120° 51′ 59.19″

[a] Types of samples: bulk precipitation (bp), fog (f), throughfall (tf), wet deposition (wd), epiphytic lichens (le), saxicolous lichens (ls), particulate matter ≤2.5 μm diameter (IMPROVE), deposition from rain and snow (NADP). Note that fog collectors were installed at Klickitat, but no data are presented because of infrequent fog events, very low fog sample volumes and vandalism of the collectors. Lichens were collected within 3 km of the coordinates.

Table 2
Precipitation at weather stations and deposition monitoring sites (See Fig. 1 for site locations) in the Columbia River Gorge

Dates	Precipitation (cm; % of total in parentheses)						
	Bonneville Dam		Hood River			The Dalles	
10/1/03 to 10/28/03 (before collection)	12.75 (7.3)		2.79 (4.9)			1.57 (4.6)	
10/29/03 to 3/9/04 (during collection)	110.39 (62.8)		39.67 (69.2)			25.25 (73.7)	
3/10/04 to 9/30/04 (after collection)	52.68 (29.9)		14.91 (26.0)			7.44 (21.7)	
2004 Water Year (Oct. 1, 2003 to Sept. 30, 2004)	175.82		57.38			34.26	
	Deposition Study Sites						
	MZ	Klick	7MH	HTSP	WI	CE	MH
10/29/03 to 3/9/04	95.23	28.95	28.17	22.03	22.16	18.08	20.36

Precipitation data from the Integrated Plant Protection Center – Degree Day Calculator (http://ippc2.orst.edu/cgi-bin/ddmodel.pl). Locations of the weather stations are as follows: Bonneville Dam: Bureau of Reclamation Agrimet; Bonneville Dam, WA – BNDW – Latitude: 45° 38′ Longitude: 121° 55′ Elevation: 24 m. Hood River: Bureau of Reclamation Agrimet station – Hood River Experimental Station Oregon – HOXO; Latitude: 45° 41′ Longitude: 121° 31′ Elevation: 174 m. The Dalles: KDLS METAR Station – The Dalles Municipal Airport, WA – DLS; Latitude: 45° 38′ Longitude: 121° 10′ Elevation: 74 m.

Bulk precipitation samples were collected with a standard rain gauge funnel (10 cm i.d.) connected to a polyethylene storage bottle. Throughfall samplers used the same funnels but directed throughfall (precipitation collected below tree canopies) to an ion exchange resin (IER) column, which captured ions as the solution percolated through the column (Fenn and Poth, 2004). After the sampling period, resin columns were extracted with 1 N potassium iodide using a modification of the method described by Simkin et al. (2004). Nitrate and sulfate concentrations in the extracts were analyzed by high performance ion chromatography (Dionex, Model DX600; Sunnyvale, CA). Slight modifications from the methods of Simkin et al. (2004) were ten-fold dilutions of samples and blanks, 9.0 mM eluent (Na_2CO_3) concentration, and a 1.0 mL min^{-1} flow rate. Ammonium was analyzed colorimetrically with a TRAACS 800 Autoanalyzer (Tarrytown, NY). At remote Twin Tunnels (TTL, TTH) and Catherine Creek (CCL, CCH), bulk deposition was also collected with IER collectors instead of weekly collection of liquid samples. Deposition fluxes calculated from IER collector data were blank-corrected using background levels from field blanks installed at each of the monitoring sites.

Fog samples were obtained with passive line collectors composed of 168 polytetrafluoroethylene (PTFE; the same resin polymer used to make Teflon) monofilament strings (Fluortek, Easton, PA), 30 cm in length and 0.4 mm in diameter, arranged in two concentric circles, 8.0 and 8.2 cm in diameter (Fenn et al., 2000). Liquid collected by the strings during fog events drained through a funnel base into a polyethylene bottle. A plastic lid (0.5 m diameter) was mounted above the fog collectors to block vertical precipitation. The passive fog collectors were continuously exposed from the sides and as a result some samples likely included wind-blown precipitation. Bulk precipitation and fog samples were collected for pH, NO_3^-, NH_4^+, and SO_4^{2-} analysis on a weekly basis. Samples were shipped overnight to the Forest Fire Laboratory in Riverside, CA for analysis. Nitrate and ammonium in bulk deposition and fog samples were measured with the TRAACS 800 Autoanalyzer; sulfate was determined with a Dionex ion chromatograph. Fog and bulk deposition pH were determined in the field using a portable pH meter (Oakton 100 Series, portable pH/mV/°C meter, Vernon Hills, IL).

To compare the pH of wet-deposition samples to bulk-deposition and fog samples, we installed an NADP wet deposition collector at Horsethief State Park (HTSP). A wet-deposition sampler was already in operation at MZ as part of the NADP network. Bulk deposition includes a small component of dry and fog deposition because the funnel is constantly exposed to the atmosphere (Mosello et al., 1988). At HTSP, we also employed Ogawa passive samplers (Bytnerowicz et al., 2002; Ogawa and Company, 1998) to monitor ambient concentrations of NO_x, NO, NH_3 and SO_2. The samplers were installed 0.4 km from the Columbia River on bluffs within 100 m of historic pictographs. Replicate samplers were generally replaced bi-weekly from July through September, 2002, every four weeks through April 2003, and bi-weekly through mid July 2003. Some of the filter disks from the SO_2 samplers were misplaced in the laboratory, and as a result, SO_2 data are not available after the mid September 2002 sampling date. It should be noted that the passive sampler monitoring at HTSP did not coincide with the throughfall, precipitation and fog sampling described above. The latter deposition measurements did not begin until October 2003.

2.2. NADP and IMPROVE data retrieval and analysis

There are 16 IMPROVE monitoring sites and 11 NADP/NTN wet deposition sites in Oregon and Washington (Fig. 1). Mean annual concentrations of ammonium nitrate and ammonium sulfate in particulate matter $\leq$2.5 μm diameter measured every third day and for the 20% of days with worst and best visibility from 2000–2003 were obtained from the IMPROVE website (http://vista.cira.colostate.edu/improve/Data/IMPROVE/summary_data.htm). Total annual wet deposition (kg ha^{-1}) of NH_4^+, NO_3^-, SO_4^{2-} and H^+ from 2003–2004 were obtained from the NADP database (http://nadp.sws.uiuc.edu/). We used Dunnett's (1955) method to compare mean concentrations of nitrogen- and sulfur-containing pollutants at CRG sites to other Oregon and Washington sites.

2.3. Lichen data collection and analyses

To indicate availability of atmospheric N- and S-containing pollutants to lichens, twenty grams each of the two most abundant lichens from a shortlist of target species were collected at each of 145 0.38-acre circular plots in the Scenic Area in summers 1998– 2004 following Geiser (2004). Target epiphytes were *Evernia prunastri* (L.) Ach., *Hypogymnia inactiva* (Krog) Ohlsson, *Letharia vulpina* (L.) Hue, and *Platismatia glauca* (L.) Culb. & C. Culb) At non-forested sites (and for comparison purposes in some forested sites), the saxicolous lichen, *Xanthoparmelia cumberlandia* (Gyelnik) Hale, was collected from boulders. All are medium-sized foliose lichens except fruticose *L. vulpina*. Because no single lichen species occurred on every plot, we collected two species to provide data overlap for assessing the validity of combining data from different species.

Epiphytes were air-dried. *Xanthoparmelia* samples were sonicated five times in one liter of distilled water for one minute then air dried to remove most of the mineral debris trapped externally between overlapping lobes. Samples were mailed to the University of Minnesota Research Analytical Laboratory (UMRAL) in St. Paul for total N and S determinations. Total N concentrations were determined with a LECO FP-528 total N analyzer and total S was determined with a LECO SC-132 S analyzer (LECO Corp., St. Joseph, MI) as described in Geiser (2004). All lichen and plant reference materials were analyzed following the quality assurance program of Geiser (2004); reference material determinations were within certified ranges.

The restricted maximum likelihood method (MathSoft, 1999) was used to estimate the variance of N and S concentration measurements due to epiphytic lichen species within plots and the variance between plots (Patterson and Thompson, 1975). The variance between plots was about twice the variance due to species within plots, and the variance between plots was similar for single species and the average for all species. Therefore values for each species

at each site were averaged to obtain an overall site average, and this final site value was used for further analysis. The saxicolous *Xanthoparmelia*, which tended to have higher N and S concentrations than epiphytes, was analyzed separately.

Lichen accumulation of N and S was compared to N and S deposition fluxes at monitoring sites located within 3 km of the corresponding lichen plots. We also regressed 52 observations (site averages) of lichen N and S concentrations from the valley floor of the Scenic Area in 2004 against longitude to look for responses to west end vs. east end pollutants. A polynomial of degree 3 provided the best fit (JMP, Version 5. SAS Institute Inc., Cary, NC, 1989–2002).

Finally, we supplemented CRG lichen data with pollution-indicator lichen and elemental data from the Oregon-Washington frame-work of Forest Service-protocol plots (USDA Forest Service, 2004; Geiser, 2004). A detailed analysis of lichen community and elemental N and S data in relation to air quality, wet deposition of ammonia, and climate in western Oregon and Washington is reported in Geiser and Neitlich (in press). We used Dunnett's (1955) method to compare mean N and S content of epiphytic lichens from the Scenic Area sites to the urban and rural-agricultural (Willamette Valley) sites, and to the bordering national forests (Mt. Hood and Gifford-Pinchot). Fig. 1 shows the location of the lichen sites. Finally, we made a bi-state map of presence/absence distributions of three lichens with strong growth responses to atmospheric sources of fixed N (Geiser and Neitlich, in press), *Candelaria concolor* (Dicks.) Stein, *Physcia adscendens* (fr.) H. Olivier, and *Xanthoria polycarpa* (Hoffm.) Rieber. All three species have broad climate and elevation ranges in the Pacific Northwest (McCune and Geiser, 1997).

3. Results

3.1. Sulfur and nitrogen deposition

Throughfall deposition was highest at Seven Mile Hill (7MH): 25.4 kg N ha^{-1} (about 50:50 NH_4^+-N and NO_3^--N) and 6.5 kg S ha^{-1} (Fig. 3). Throughfall deposition of NO_3^- was greater than NH_4^+ at sites west of 7MH. To the east of 7MH throughfall deposition of NH_4^+ was greater than or similar to NO_3^- deposition except at Klickitat (KLICK), a site located to the north and outside of the main drainage of the CRG. Throughfall deposition of NH_4^+ was greater at sites east (mean 8.01 kg N/ha) than west (mean 2.76 kg N ha^{-1}) of 7MH (Table 3). The east:west ratios of throughfall NO_3^-, NH_4^+, and SO_4^{2-} were 1.16, 2.90 and 1.36.

Unlike throughfall deposition, bulk deposition of NO_3^-, NH_4^+, and SO_4^{2-} was higher west than east of 7MH. Bulk deposition was generally similar at all the sites west of 7MH, except that NH_4^+ deposition at MZ was about half that of the other western sites (Fig. 3). Bulk deposition values were also similar among sites east of 7MH, but values were consistently lower than at 7MH and sites west of 7MH (Fig. 3). The east:west ratios of NO_3^-, NH_4^+, and SO_4^{2-} in bulk deposition were 0.74, 0.54 and 0.67 (Table 3).

Although the fog collectors were not designed to exclude wind-blown precipitation, they appear to have mostly sampled fog, as evidenced by the lower volumes and higher ionic concentrations of the fog collector samples compared to the bulk deposition samples (Fig. 4). However, at MZ and 7MH, high volumes in the fog collectors on many dates (and consistently low ionic concentrations at MZ) suggest that wind blown precipitation diluted some samples. Contamination of fog samples by rain or snow was probably minor in sites other than MZ and 7MH. At KLICK and CE fog samples were only collected on one sampling date and the volume was extremely low at KLICK (Table 4).

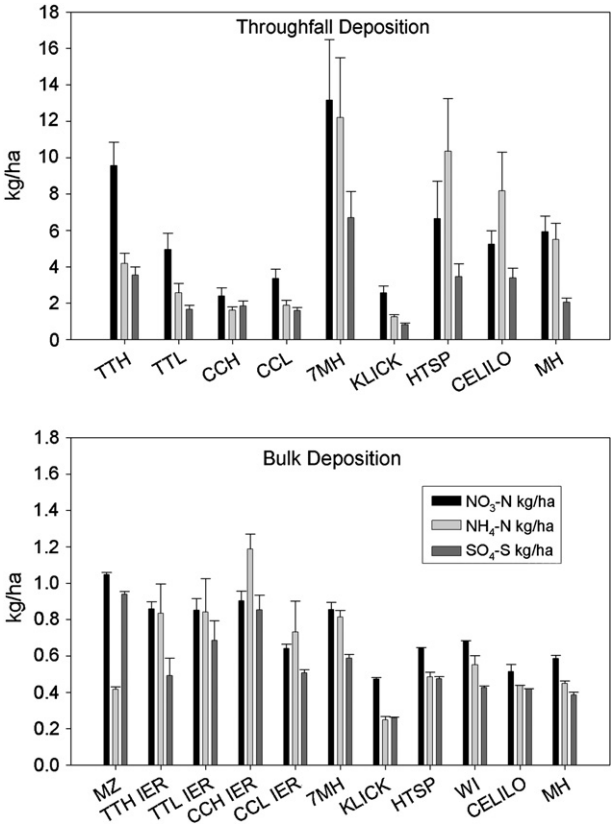

Fig. 3. Throughfall and bulk deposition fluxes during the 19 week monitoring period (October 2003 to March 2004). Error bars represent standard errors of the mean.

At 7MH NO_3^- and NH_4^+ concentrations and cumulative deposition fluxes in fog (mg m^{-2}) were highly similar and were

Table 3
Comparison of nitrogen and sulfur deposition (kg ha^{-1}) in throughfall and bulk deposition at sites located in the eastern versus western portions of the main study area

Ion	Western Sites	Eastern Sites	East:West Ratio
	Throughfall Deposition		
NO_3-N	5.11	5.94	1.16
NH_4-N	2.76	8.01	2.90
SO_4-S	2.18	2.97	1.36
	Bulk Deposition		
NO_3-N	0.81	0.61	0.74
NH_4-N	0.90	0.48	0.54
SO_4-S	0.63	0.43	0.67

Western sites include Twin Tunnels high and low elevation and Catherine Creek high and low elevation sites. Eastern sites include Horsethief State Park, Wishram (bulk deposition only), Celilo and Maryhill. The Seven Mile Hill site was not included in these west-to-east comparisons because this site is higher in elevation and more exposed to fog deposition than the other sites and because this site is located approximately midpoint between the western and eastern sites. Data from the Mt. Zion site are not included either, because this is an extremely western site and strongly influenced by the Portland urban pollution plume. The Klickitat site was also excluded from the east/west comparison because it is located within a separate side drainage north of the main gorge and may not be exposed to the principal air masses coming through the gorge to the same extent as the other sites.

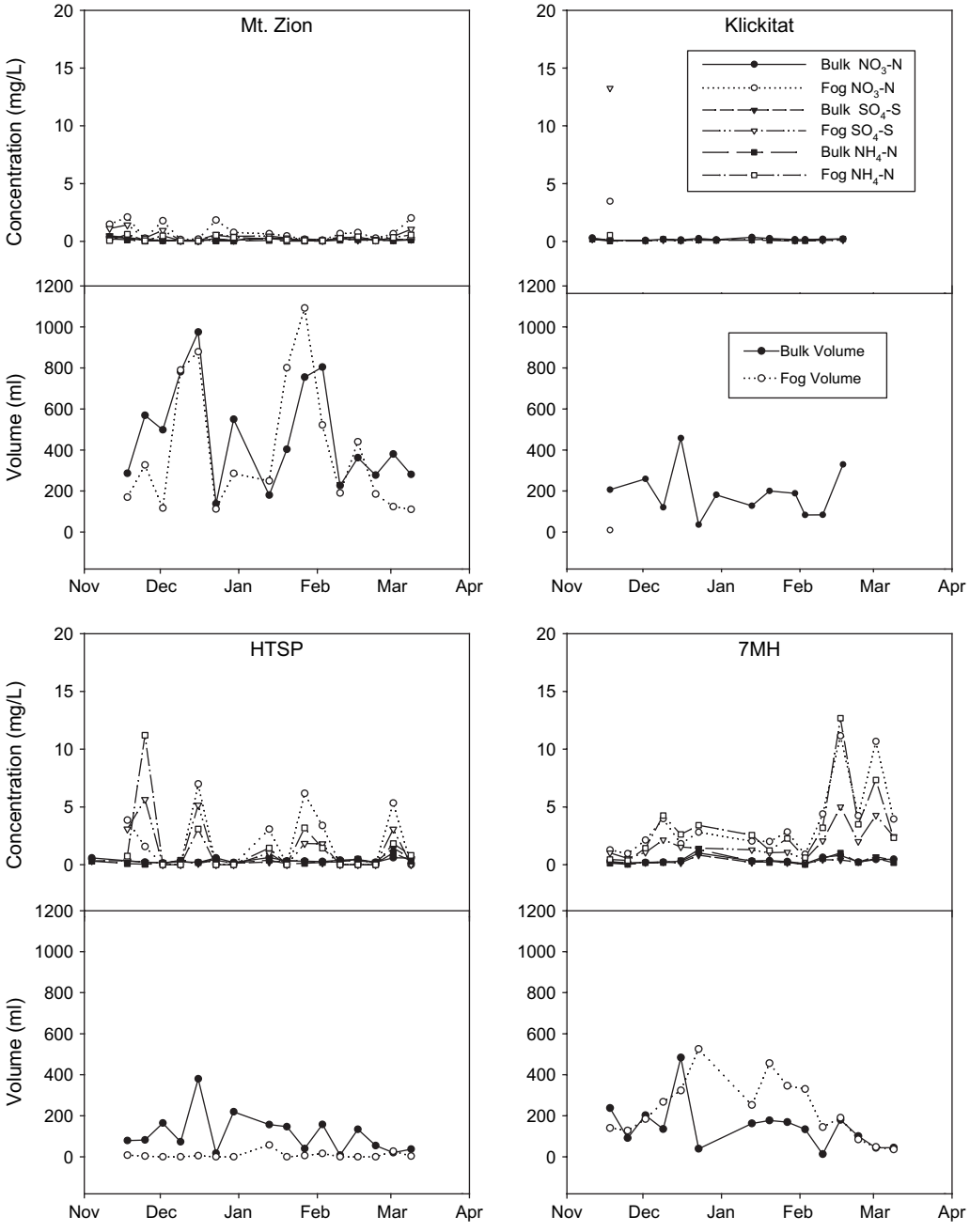

Fig. 4. Concentrations of NO_3^-, SO_4^{2-} and NH_4^+ in bulk deposition and fog samples.

greater than sulfate concentrations (Fig. 3) and deposition (Table 4). These results parallel the throughfall data from 7MH, suggesting that fog deposition to tree canopies is an important source of throughfall fluxes in the CRG. Cumulative deposition fluxes of N and S in fog to the string collectors during the monitoring period (mg m^{-2}) were 2–3 times higher at 7MH than fluxes at the next highest sites (WI and MZ), and were as much as two or three orders of magnitude higher than fog deposition at HTSP, CE and KLICK (Table 4). At all the sites except 7MH, NO_3^- concentrations and deposition fluxes in fog were higher than for NH_4^+ or SO_4^{2-}. At the two eastern Gorge sites with frequent fog occurrence (7MH and WI) ionic concentrations and deposition fluxes decreased in the order $NO_3^- \geq NH_4^+ > SO_4^{2-}$. At MZ, the only site on the western end of the Gorge, SO_4^{2-} concentrations and deposition were higher than for NH_4^+ but lower than NO_3^- (Table 4).

3.2. Deposition acidity

The pH of fog and precipitation varied temporally at all sites. A few pH values were ≥5.7, particularly in November and early December. The most acidic samples were collected at WI in January (Fig. 5). Fog samples from 7MH were the most consistently acidic; December–March pH was 3.8–5.0 and was <4.5 on 9 of 12 sampling dates. Bulk precipitation pH at 7MH was ≥4.5. Precipitation or

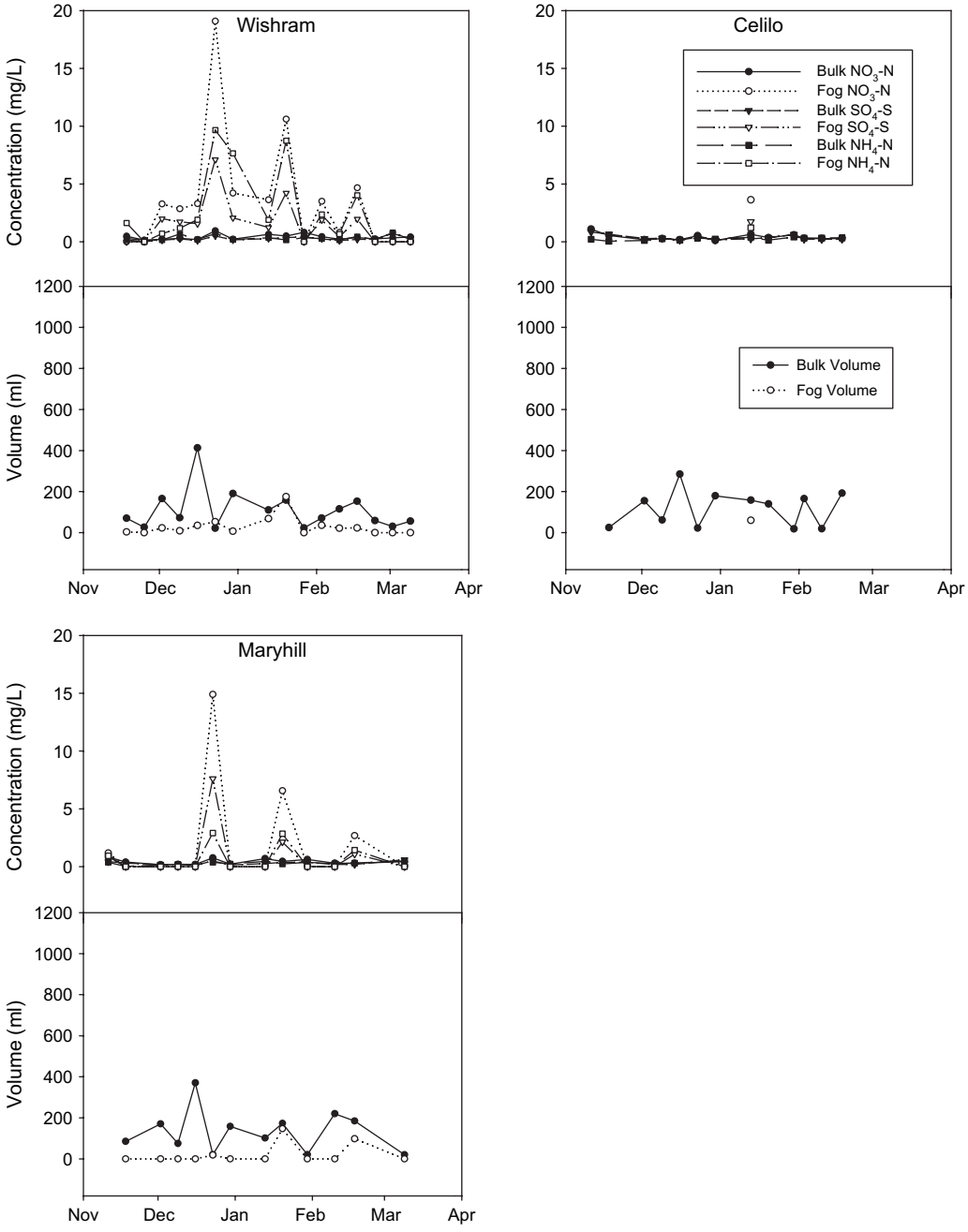

Fig. 4. (*continued*)

fog pH values ≤4.5 occurred at KLICK (4 dates), HTSP (2 dates), CE (3 dates), WI (3 dates), and MH (2 dates) (Fig. 5). At MZ, the pH of bulk fog and bulk deposition samples was 5.0–6.0 except on five dates when fog pH was 4.5–5.0 and once when fog pH was 6.4–6.6. Lower pH values at 7MH, KLICK, HTSP, CE, WI and MH, but not MZ, were more often observed during the latter part of the study, beginning in mid January.

Mean fog pH at MH varied widely on three dates (6.7, 4.4 and 5.6) and was generally less acidic than bulk precipitation pH (Fig. 5). On the 1–3 dates when fog pH was measured at HTSP, CE and MH, fog was less acidic than bulk precipitation. Fog samples were usually more acidic than bulk precipitation at MZ, 7MH and WI, the sites with greatest fog frequency and ionic fog deposition (Table 4).

3.3. Gaseous pollutants in ambient air

At Horsethief State Park, mean monthly concentrations of ambient NO_x, NO_2, NO, and NH_3 peaked between October and February at 15.3, 10.3, 9.4, and 3.7 ppb, respectively (Fig. 6). Lowest concentrations occurred in summer. In contrast to HTSP deposition data, ambient NO_x was always much higher than NH_3; ratios of NO_x:NH_3 ranged from 4–22. Summertime SO_2 was low, ranging from 0.5 to 3 ppb. Unfortunately, we do not have data on SO_2 concentrations during

Table 4
Cumulative fog deposition fluxes to passive string collectors (mg m^{-2}) at seven sites in the Columbia River Gorge

Site	NO_3-N	NH_4-N	SO_4-S	Average fog volume (ml)	No. of fog collection dates
Seven Mile Hill	162.42	157.05	83.10	230.2	15
Wishram	56.78	39.55	22.84	41.1	11
Mount Zion	46.83	16.48	29.42	399.5	17
Maryhill	23.86	9.70	8.93	88.0	4
Horse Thief State Park	4.81	1.85	2.78	17.5	7
Celillo	3.44	1.15	1.65	60.0	1
Klickitat	0.49	0.08	1.88	9.0	1

winter or spring, although it seems likely that SO_2 concentrations also peaked in winter based on the temporal trends for NO_x, NO_2, NO, NH_3 and H_2S (H_2S data not shown).

3.4. Regional comparisons of particulate and wet deposition chemistry

Means comparisons using Dunnett's Test ($\alpha = 0.05$) and the Scenic Area NADP site at MZ as control, provided evidence that annual wet ammonium and inorganic-N deposition (kg ha^{-1}) at MZ were higher than all ten of the other Oregon and Washington NADP sites, even though only three sites had lower precipitation. Nitrate deposition at MZ was comparable to the deposition monitor closest to Seattle (in Marblemount, WA) and higher than other sites. MZ sulfate deposition was intermediate to and hydrogen ion concentrations were not different than other NADP sites. Mean annual wet deposition fluxes of NH_4-N, NO_3-N, SO_4-S, and H^+ were 1.2, 1.2, 1.5 and 0.07 kg ha^{-1}, respectively.

Average annual ammonium nitrate concentrations in particulate matter during 2003–2004 were higher at WI than all 15 other Oregon and Washington IMPROVE sites during the 20% of days with the worst visibility. Ammonium nitrate concentrations at WI and the IMPROVE site in the western Scenic Area (MZ) were higher than all other sites except the Seattle urban area IMPROVE site when all days were considered. Ammonium sulfate concentrations at the CRG IMPROVE sites on the 20% of worst days was intermediate to other regional IMPROVE sites; but with all days considered, were higher than all regional sites except Seattle. Average annual concentrations of ammonium nitrate in fine particulates for the 20% of worst visibility days and for all days were 4.47 and 1.25 μg m^{-3} at WI and 2.43 and 0.97 μg m^{-3} at MZ. Average annual concentrations of ammonium sulfate on the 20% of worst visibility days and 100% of days were 2.04 and 1.31 μg m^{-3} at WI and 2.35 and 1.37 μg m^{-3} at MZ.

3.5. Nitrogen and sulfur concentrations in lichen thalli

Lichen N and S accumulation patterns matched throughfall deposition patterns (Fig. 7). Highest N and S concentrations in the saxicolous lichen, *Xanthoparmelia cumberlandia*, occurred near the four easternmost stations; highest N concentrations in epiphytes (1.31%) occurred near 7MH (*X. cumberlandia* was not collected here) and lichen N concentrations were higher at the TT (1.05%) than CC sites (0.88%). High thallus N was also observed in the west at MZ (1.25%; no throughfall deposition data).

The third degree polynomial fit of the regression of longitude along the valley floor on lichen N concentrations provided evidence of higher N accumulation in lichens towards the east and west boundaries, closer to major emission sources ($r^2 = 0.33$, $F = 7.76$, $p = 0.0003$; Table 5, Fig. 8). Nitrogen concentrations decrease with distance eastward from the Portland/Vancouver metropolitan area, then increase again with proximity to the eastern border of the Scenic Area. Sulfur concentrations were similar throughout the Scenic Area (data not shown). Mean N and S concentrations of epiphytic lichens collected from the Scenic Area (1.02% N and 0.107% S) were double the concentrations in lichens from bordering Mt. Hood and Gifford Pinchot national forests (0.44–0.52% N and 0.050–0.056% S), comparable to the agriculture-dominated Willamette Valley (1.02% N and 0.110% S) and lower than urban forests (1.36% N and 0.122% S; Fig. 9).

3.6. Lichen communities

The distribution of epiphytic nitrogen-loving lichens corresponded to regional locations with high N deposition and high N concentrations in lichen thalli. Nitrophytes were detected throughout the CRG (Fig. 10). They were also frequently detected in the greater metropolitan areas of the regions largest cities: Seattle, Portland, Spokane, the Tri-cities and Salem, and in forests rimming the Columbia basin and Oregon's agricultural heartlands in the northeast, southwest, and Willamette Valley (the latter extends south from Portland and Salem between the Coast and Cascade ranges in Oregon).

4. Discussion

4.1. Atmospheric deposition levels in the Scenic Area

Atmospheric N deposition in ponderosa pine throughfall during the 19 week monitoring period was high throughout the Scenic Area (11.5–25.4 kg ha^{-1}). Throughfall NO_3^- and NH_4^+ fluxes show that both oxidized and reduced forms of N were important fractions of total inorganic N deposition. The highest deposition fluxes of NH_4^+ in throughfall were measured at 7MH and eastward, consistent with emissions data. Elevated NO_3^- deposition is typical of areas exposed to emissions from fossil fuel combustion, especially from transportation and industrial sources. Elevated NH_4^+ deposition typically indicates agricultural sources, such as confined animal feeding operations, although emissions from mobile, sewage and industrial sources can also contribute (US EPA, 2005).

Deposition to open or canopy-free areas was much lower than deposition under canopies because tree surfaces collect fog and dry deposition. Thus, deposition over the larger landscape depends on canopy density, edge effects, and canopy surface area (Weathers et al., 2001). Bulk deposition in this study was always several-fold lower than throughfall

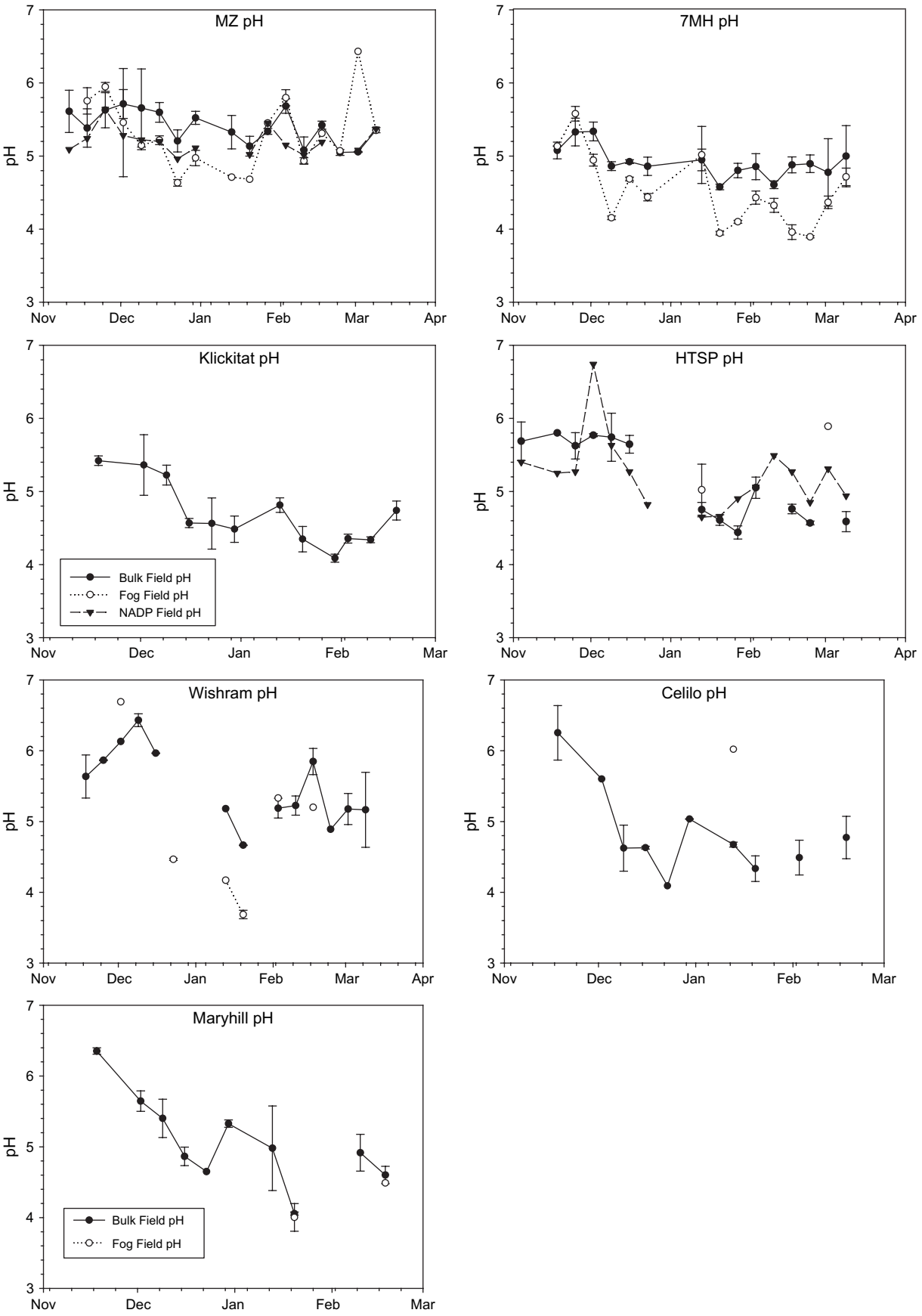

Fig. 5. The pH of fog or bulk precipitation samples. The pH data for the NADP type sampler at HTSP and MZ are also included.

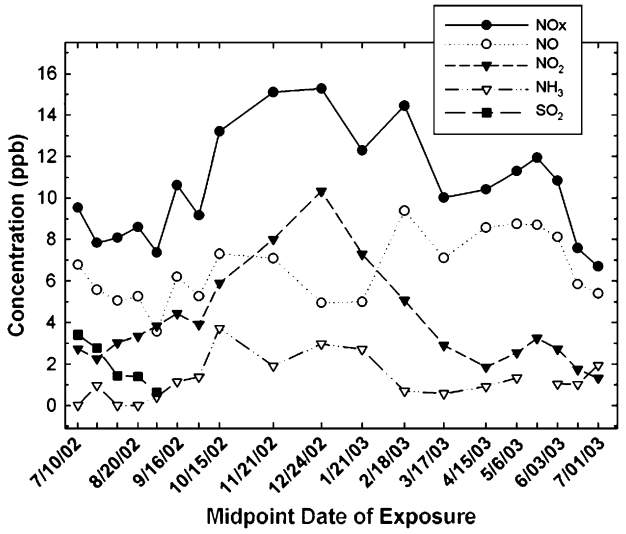

Fig. 6. Average ambient concentrations of NO_x ($NO_2 + NO$), NO_2, NO, NH_3, and SO_2 measured with passive samplers at Horsethief State Park from July 2002–June 2003.

deposition, demonstrating the greater importance of fog and dry deposition to these forests compared to wet deposition in rain or snow. This is because ionic concentrations are many-fold higher in fog than in rain or snow and because dry deposition to forest surfaces is a continuous process during precipitation-free periods. Thus, throughfall deposition provides an estimate of total deposition (wet, dry and fog deposition); however in the case of N deposition, throughfall provides a lower-bound estimate of total deposition because of canopy consumption of N (Lovett and Lindberg, 1993).

Deposition of NO_3^- in throughfall was usually much higher than deposition of SO_4^{2-}, and in the eastern sites, NH_4^+ deposition was also much higher than SO_4^{2-} deposition. Nonetheless, S deposition was elevated compared to unpolluted regions. Sulfur deposition in throughfall at 7MH was 6.7 kg ha^{-1} during the 4.5 month monitoring period and ranged from 0.8 to 3.5 kg ha^{-1} at the other throughfall sites (Fig. 3). Bulk deposition of SO_4^{2-} was greatest at MZ, the site with the highest precipitation, and bulk deposition of NO_3^- and SO_4^{2-} at MZ were highly similar

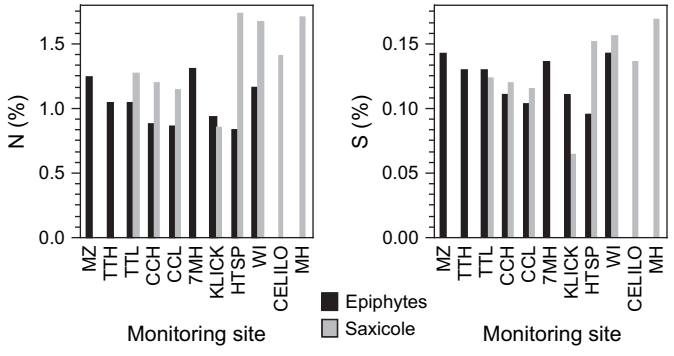

Fig. 7. Concentrations of nitrogen and sulfur in epiphytic and saxicolous lichens collected within 3 km of deposition monitoring sites in the Columbia River Gorge from 1998–2004.

Table 5
Parameter estimates for the 3° polynomial fit of 52 lichen thallus % N observations regressed on longitude, CRG valley floor, 2004

Term	Estimate	Std Error	*p*-value
Intercept	65.1798	19.676	0.0013
Longitude	0.525	0.1618	0.0016
Longitude^2	0.4764	0.1739	0.00125
Longitude^3	−0.85	0.4431	0.0516

(1.05 and 0.94 kg ha^{-1}, respectively). Throughfall wasn't collected at MZ because it is a canopy-free NADP monitoring site. Sulfate deposition in fog was nearly three-fold greater at 7MH than at MZ, but fog deposition of SO_4^{2-} was highly similar at MZ and WI (Table 4), suggesting that SO_4^{2-} deposition may be relatively similar at eastern and western Gorge sites. Throughfall deposition of SO_4^{2-} was also similar at both ends of the Gorge. These findings are in agreement with the documentation of elevated S concentrations in lichens throughout the Scenic Area.

We sampled fog, bulk deposition and throughfall in the Scenic Area during the major portion of the wet season. Precipitation levels during our sampling period corresponded to 69–74% of the annual precipitation (Table 2) at Hood River and The Dalles for the 2004 water year (Oct 2003 to September 2004; http://ippc2.orst.edu/cgi-bin/ddmodel.pl). Although throughfall fluxes were relatively high during this 19 week study, annual inputs, especially to forested areas, were probably much higher because dry deposition during summer was not measured in this study. The first storm of the wet season, equaling 4–5% of the annual precipitation total, occurred in October just before sampling began. Because we did not sample the initial rain event, we likely underestimated throughfall and bulk deposition. Throughfall deposition may have been particularly underestimated because the first precipitation event of the water year is likely to have resulted in a flush of N and S ions that accumulate on canopy surfaces over the summer dry season. The length of the antecedent dry period between rain events can be an important factor affecting throughfall deposition fluxes (Lovett and Lindberg, 1984). At Hood River and The Dalles meteorological stations (Fig. 1), only 1.42 and 0.23 cm of precipitation were recorded from June–September, 2003, suggesting that the first throughfall deposition event contained higher N and S levels compared to subsequent rain events with short antecedent dry periods.

Peak concentrations of gaseous N and S pollutants at HTSP were indicative of a diluted urban plume (Bytnerowicz and Fenn, 1996; Bytnerowicz et al., 1998), or in the case of NH_3, agricultural emissions were likely an important source. The low to intermediate gas concentrations at HTSP, but high throughfall deposition of NO_3^- and NH_4^+ and moderate SO_4^{2-} deposition, suggest that S- and N-containing air pollutants are primarily transported to HTSP in particulate rather than gaseous form. However, nitric acid vapor (HNO_3) concentrations were not measured and could be an important N deposition input in areas affected by the urban plume from the western end of the Gorge. The high

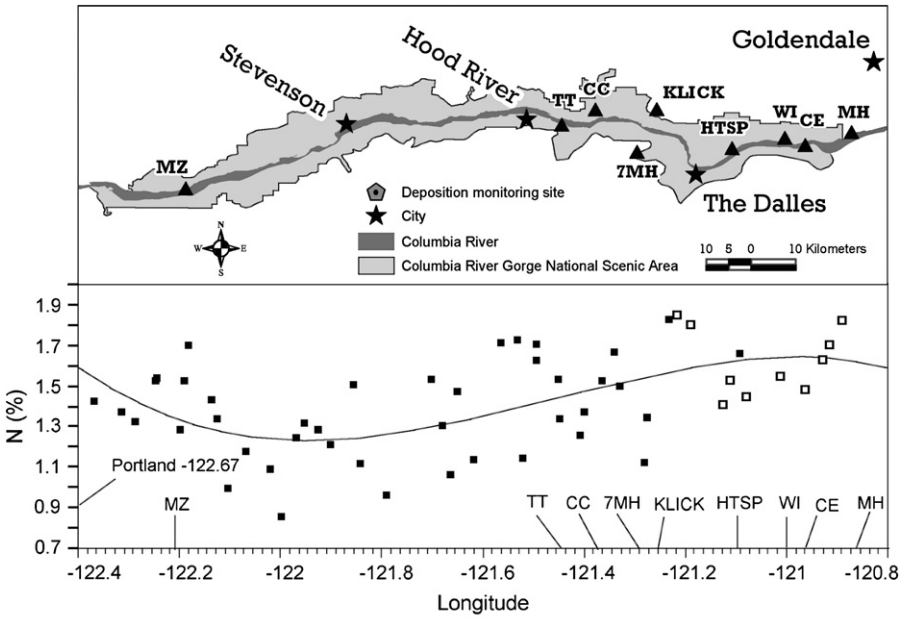

Fig. 8. Nitrogen concentrations (% dry weight) in lichens collected from west to east along the Columbia River Gorge National Scenic Area valley floor in 2004. Epiphytic lichens: closed squares; saxicolous lichens: open squares. Short lines point to the corresponding longitude of Scenic Area deposition, NADP and IMPROVE monitors (Table 1).

NO_x:NH_3 ratio at HTSP also suggests the possibility that a higher proportion of NH_x than of NO_x is deposited to plant surfaces and washed off in throughfall events, although this requires further study.

4.2. Spatial trends in deposition

East of 7MH, the throughfall deposition pattern was generally $NH_4^+ \geq NO_3^- > SO_4^{2-}$, consistent with greater agricultural emissions in the eastern CRG. Higher concentrations of NH_4^+ deposition in throughfall and N concentrations in lichens from 7MH eastward suggest higher fog and/or dry deposition of NH_4^+ in the eastern Scenic Area. The primary NH_3 sources within and to the east of the CRG Scenic Area are livestock operations related to beef and dairy production, and crop fertilizer applications (US EPA, 2005).

West of 7MH, throughfall NO_3^- concentrations were higher than NH_4^+ or SO_4^{2-} concentrations and NH_4^+ concentrations tended to be similar to or slightly greater than SO_4^{2-} concentrations. This reflects the high NO_x emissions profile of the urban source area (greater Portland), which is the major influence on the western end of the gorge. Emissions from the west end are primarily from commercial marine vessels and highway vehicles, with significant contributions from off-highway vehicles (notably rail traffic and construction), and fuel combustion by industry (US EPA, 2005). Bulk deposition of NO_3^-, NH_4^+ and SO_4^{2-} was higher west of 7MH, presumably because precipitation is much greater in the western end of the Scenic Area. Higher N accumulation in lichens toward the western and eastern boundaries are further evidence of the influence of neighboring county emissions.

The highest N concentrations in lichen thalli in the eastern CRG correspond with high N deposition in throughfall and fog in the eastern CRG. Lichens absorb dry-deposited solutes when moistened and can directly absorb solutes in precipitation and fog, hence they respond to both wet and dry deposited forms of N and S. Nitrogen and S concentrations were generally higher in *X. cumberlandia* than in epiphytes from the same sites, which we attribute to the formation of over-lapping lobes that trap fine soil and dust particles, despite the surface rinsing procedure we applied.

4.3. Potential effects of deposition on cultural resources

Precipitation in equilibrium with CO_2 has a pH of 5.65, slightly acidic from carbonic acid formed by CO_2 dissolution in water. Precipitation of pH 5.65 provides a benchmark for evaluating pollutant influences on precipitation and fog acidity. Fog and bulk precipitation samples with pH $\leq$ 4.5 on two or more sampling dates at all monitors east of MZ demonstrate that acidic deposition occurs throughout the eastern Scenic Area. Fog and bulk precipitation was never <4.5 at MZ. More extreme acidic events, here defined as pH < 4.0, occurred on three sampling dates at 7MH and once at WI and MH, and additional low pH events might have been detected if the fog collectors had reliably excluded wind-blown precipitation.

Acidic deposition detrimentally affects buildings and monuments of certain rock types (e.g., marble) and, where stones remain wet for long periods, can be a significant or primary cause of damage (Charola, 2001; NAPAP, 1998). Episodic acidity from precipitation and fog in the CRG poses an as yet unquantified risk to rock paintings and carvings, which

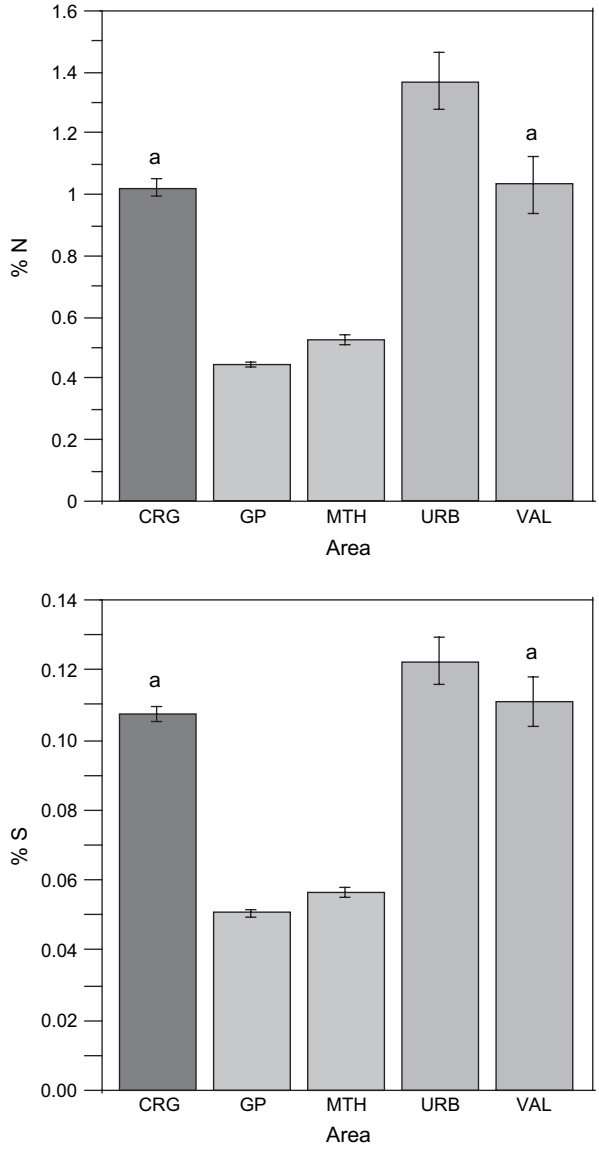

Fig. 9. Mean epiphytic lichen N and S concentrations (% dry weight) in the Scenic Area (CRG, $n = 145$) and adjacent national forests to the north (GP: Gifford-Pinchot, $n = 110$) and south (MTH: Mt. Hood, $n = 112$), urban areas (URB, $n = 29$), and rural forests of Oregon's Willamette Valley (VAL, $n = 9$). Lowercase letters above bars indicate areas that are not different from the CRG. Error bars indicate standard error. Samples were collected in summer, 1996–2004.

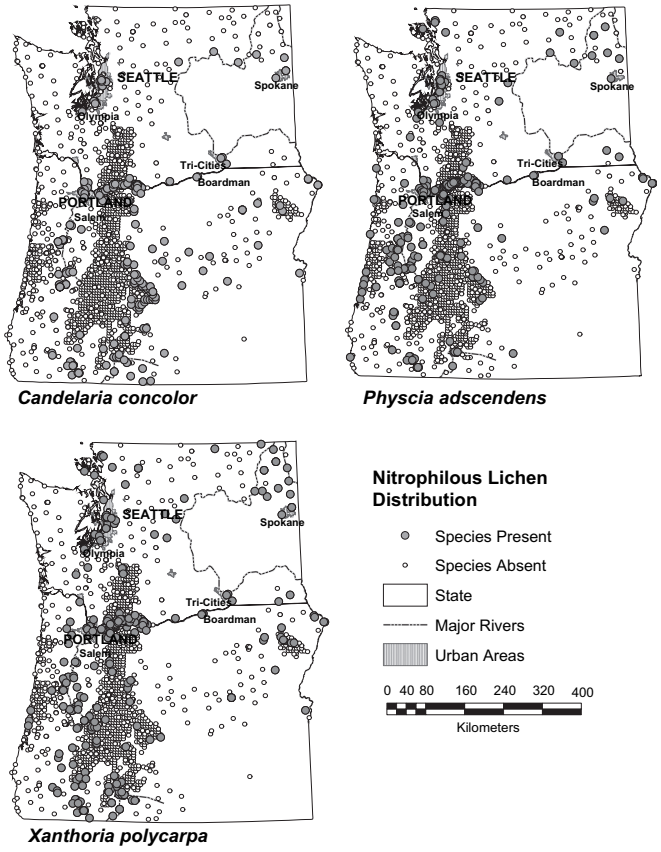

Fig. 10. Distribution of the nitrophilous, epiphytic lichens: a) *Candelaria concolor*, b) *Physcia adscendens*, and c) *Xanthoria polycarpa* in the Columbia River Gorge environs. Lichen sampling only occurred in forested areas.

occur mostly on basalt in relatively dry environments. Nitrogen and S deposition may also pose a threat to these cultural resources. Eutrophication stimulates the growth of natural surface dwelling algae and sulfur-, and nitrogen-oxidizing chemotrophic bacteria. Such bacterial growth weathers cultural stone by releasing nitric and sulfuric acids and by the physical swelling and shrinking of microbial biomass with moisture and temperature changes (Mansch and Beck, 1998; Viles, 1995). Ammonia gas can directly corrode clay pigments used in rock paintings. However, ambient NH_3 concentrations were low at HTSP, while ammonium nitrate and ammonium sulfate concentrations in particulates detected by IMPROVE, and wet and bulk deposition of NH_4^+, were high compared to other parts of the bi-state region and compared to background levels for remote sites. These findings suggest that a large proportion of the N detected in deposition and lichen thalli came from long distance transport of ammonium nitrate and ammonium sulfate particles.

4.4. Potential effects of deposition on natural resources

Soil acidification and eutrophication (N-enrichment) are the most common ecosystem responses to chronic N and S deposition in western North American forests (Fenn et al., 2003). Lichens are sensitive indicators of N enrichment, and the lichen studies in the CRG clearly show that ecological effects from air pollution are occurring. Lichen communities in many areas of the CRG, notably the valley floor, support enhanced populations of nitrogen-loving species (Geiser and Neitlich, in press), and lichen N concentrations are elevated relative to neighboring forests to the north and south and comparable to agriculturally-intense environments. Chronic deposition of 10–25 kg N ha^{-1} yr^{-1} as reported in this study is within the range at which N fertilization responses and symptoms of N excess or N saturation have been reported in European forests (Dise and Wright, 1995), in eastern North America (Aber et al., 2003; Peterjohn et al., 1996; Stoddard, 1994), and in chaparral and forested watersheds in southern and central California (Fenn and

Poth, 1999; Fenn et al., 1998, 2003). Estimated deposition inputs as low as 7.5 kg ha^{-1} yr^{-1} in high elevation catchments in the Colorado Front Range has clearly resulted in N saturated conditions as evidenced by elevated NO_3^- concentrations in surface runoff (Fenn et al., 1998, 2003; Williams et al., 1996) and in high elevation lakes (Musselman and Slauson, 2004). Enhanced accumulation of N in the ecosystem leads to the syndrome of ecosystem responses referred to as N saturation (Aber et al., 1989; Fenn et al., 1998). Symptoms of N saturation include reduced C:N ratios in mineral and organic soil layers and increased foliar N, soil acidification, N cycling and N mobilization. Excess N stimulates microbial nitrification, the key process causing increased nitrate leaching and gaseous emissions of N from soils (Fenn et al., 1998). Additional ecosystem effects reported in western North America as a result of excess N include increased tree susceptibility to insects and diseases (Jones et al., 2004), altered plant shoot: root ratios, decreased diversity of mycorrhizal communities, and enhanced growth of invasive species, among others (Fenn et al., 2003).

Soil acidification is a gradual process caused by both N and S deposition. As NO_3^- and SO_4^{2-} leaching increases, leaching of counter-balancing base cations also increases, amplifying the acidifying effects of nitrification. Ecosystem risks from soil acidification depend on soil buffering capacity. Most semi-arid soils are well buffered and of relatively high soil base cation saturation compared to highly leached mesic soils. Based on this short-term study, the highest deposition inputs occur in the eastern CRG, where precipitation and base cation leaching potential is lower than in the western end of the Gorge. Thus, N enrichment effects are of more particular concern than soil acidification effects in the semiarid forests of the eastern CRG Scenic Area where N deposition levels are as high as 25 kg ha^{-1} yr^{-1}. Further study is needed to ascertain the risk of N saturation impacts and soil acidification throughout the Scenic Area.

Phytotoxic effects to vascular plants from SO_2 exposure, if present in the CRG, are likely only in the vicinity of a large point source. Unfortunately, much of the SO_2 data from HTSP in this study was lost, but considering that some lichen species native to the Pacific Northwest are sensitive to SO_2 concentrations as low as 5–10 ppb (Deruelle, 1978; DeWit, 1976; Van Dobben and ter Braak, 1999), further studies of SO_2 effects on lichen communities in the CRG environs is warranted. Sulfur enrichment of lichen thalli throughout the CRG (Geiser and Neitlich, in press) is extensively documented, but the physiological effects and impacts of S deposition on lichen communities in the CRG are not known, although acidic deposition has been shown to be harmful to lichens (Farmer et al., 1992).

4.5. Emissions sources and prognosis for reductions

Where do Scenic Area pollutants originate and what can be done to mitigate them? Primary source regions are the Scenic Area itself, the Columbia Basin (including counties adjacent to the Scenic Area along the Columbia River to the east), and the Portland/Vancouver metropolitan areas to the west. The relative contributions from each region vary seasonally with summer (westerly), winter (easterly) and transition (variable) wind patterns, precipitation and topographic factors.

Scenic Area emissions are dominated by transportation, while other Scenic Area emission categories are minor compared to Basin and urban emissions. In winter, emissions from the Columbia Basin facilitate ammonium nitrate and ammonium sulfate formation transported to the Scenic Area by moist easterly winter winds. Important Basin sources include the co-located Boardman coal-fired power plant, emitting 72% and 15% of Basin SO_2 and NO_x, and the Three Mile Canyon factory farm (53 000 cattle, scheduled to increase to 90 000), and various other sources of NH_3 and NO_x. CALPUFF modeling results (John Vimont 2005; personal communication) show the Boardman coal-fired power plant by itself contributing an average of 64 days each winter to visibility impairment in the Scenic Area—a high frequency even considering modeling uncertainty. Emission controls on the power plant and large factory farms are feasible and stricter emission controls for the power plant may occur within 5–7 years. Major NH_3 sources (feedlots, dairy farms, crop fertilizer application and smaller livestock operations) are unregulated, yet contribute more than 40% of the total N emissions in the combined source regions shown in Fig. 2.

Summer westerlies favor transport of urban pollutants, primarily from mobile sources (e.g. trucks, cars, marine vessels, trains, construction and farm equipment) and industry toward the Scenic Area. Higher summer wind speeds and drier weather disperse pollutants more rapidly, moderating net transport. New EPA and Oregon State initiatives, when enacted, will phase in cleaner fuels for most transportation sources. All west coast states have adopted California CO_2 tailpipe standards which reduce N emissions commensurately. High fuel prices will push consumers toward lower emission vehicles. Marine vessels at international ports in Portland, Vancouver and Longview contribute 28% of NO_x and 12% of SO_2 emitted by this tri-county urban source region, a proportion comparable to the hundreds of thousands of urban highway vehicles. Emission reductions for marine traffic are needed, but regulation is slow because of the international aspect of commercial shipping. Using shore power while in port and cleaner fuels, would significantly reduce emissions.

5. Conclusions

We documented N and S deposition and fog/precipitation acidity in the Columbia River Gorge National Scenic Area at levels known to adversely affect natural resources and likely to affect cultural resources to some degree, although this has not been studied in the CRG. Atmospheric samples, lichen thalli, and fine particulates from the Scenic Area contained higher concentrations of N than other regional sites. Nitrogen-loving lichens, indicative of a eutrophied environment, were common throughout the Scenic Area. Forested areas may be at greatest risk for eutrophication and soil

acidification due to higher depositional surface area and precipitation. Spatial patterns of NH_4^+ and NO_3^- deposition were consistent with NO_x-dominated urban emissions in the west end and NH_3-dominated emissions in the rural east end of the Gorge. Low emissions and ambient gas concentrations in the Scenic Area itself suggest that dry deposition of gaseous pollutants is not the major deposition form in the eastern CRG. Rather, high N concentrations in fog, wet deposition and aerosol samples, indicate that transported particulates and subsequent dry deposition along with deposition in precipitation and fog, account for much of the N and S enhancement in the Gorge.

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