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Throughfall deposition of nitrogen and sulfur in a Jeffrey pine forest in the San Gabriel Mountains, southern California

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Abstract

Deposition of N and S in bulk precipitation and throughfall, and dry deposition to surrogate surfaces were determined in 1994 in a high-elevation mixed conifer forest dominated by Jeffrey pine (*Pinus jeffreyi* Grev. & Balf.) in the San Gabriel Mountains northeast of Los Angeles, California. Three sites 2 km apart, with a range of N deposition inputs, were chosen for study. N deposition in throughfall and dry deposition fluxes of N varied among the three sites, presumably because of differing pollutant transport with varying air flow patterns as a function of ridge and canyon orientation and the prevailing northeasterly winds. Net throughfall, an estimate of dry deposition, was the dominant input source of NH_4^+ and NO_3^-, except at Vista where wet deposition of NH_4^+ was greater than dry deposition of NH_4^+. Wet deposition of SO_4^{2-} was much greater than dry deposition of SO_4^{2-} at all three sites. Higher SO_4^{2-} deposition in bulk precipitation and throughfall in this study compared to montane sites further from the coast was presumably because of marine aerosols transported inland during storm events and possibly by the prevailing winds. The location of the study sites in relation to the ocean (65 km inland) and to the urban smog source, resulted in throughfall inputs which were higher in S (7.8–12.5 kg ha^{-1} $year^{-1}$), but lower in N (6.0–11.5 kg ha^{-1} $year^{-1}$) compared to montane sites in the lower San Gabriel Mountains (23 kg N ha^{-1} $year^{-1}$) or in the western end of the San Bernardino Mountains further inland (19 kg N ha^{-1} $year^{-1}$; 3 kg S ha^{-1} $year^{-1}$). S concentrations in pine foliage (0.69–0.83 g kg^{-1}) were also greater in the San Gabriel Mountain sites than in four sites located further inland in the San Bernardino Mountains (0.48–0.56 g kg^{-1}). Results of this and other studies suggest that the threshold for throughfall N deposition levels which lead to symptoms of N saturation in southern California montane ecosystems is ca. 16–19 kg ha^{-1} $year^{-1}$.

Keywords: Atmospheric deposition; Marine aerosols; Seasalt sulfate; Sulfur accumulation; Dry deposition; Surrogate surfaces; Nitrogen deficiency

1. Introduction

N deposition in terrestrial ecosystems located downwind of Los Angeles and in adjacent urban areas is among the highest in North America. By comparison, anthropogenic S deposition is relatively low compared to northeastern North America (Bytnerowicz and Fenn, 1996). Mixed-conifer forests in the western high-pollution end of the San Bernardino Mountains and chaparral stands in the San Gabriel Mountains (SGM) near Los Angeles exhibit symptoms of N saturation (Aber et al., 1989; Fenn et al., 1998). The primary symptoms of N saturation in these sites include: high streamwater fluxes of NO_3^-, elevated soil solution NO_3^-, high foliar N:P ratios, foliar accumulation of NO_3^-, enhanced nitric oxide (NO) emissions, low soil C:N ratios, and lack of a growth response to N fertilization (Anderson et al., 1988; Fenn et al., 1996; Kiefer and Fenn, 1997; Riggan et al., 1985).

N deposition in low-elevation chaparral watersheds in the SGM is high, due to proximity to the major N sources and location below the atmospheric inversion layer (Riggan et al., 1985; Bytnerowicz et al., 1987a; Solomon et al., 1992). However, few data are available on N and S deposition in higher elevation mixed-conifer forests in the SGM. Atmospheric deposition in the SGM is expected to decrease with elevation due to simple dilution effects. Deposition is also expected to vary based on local topographic characteristics influencing exposure to pollutant transport from incoming air masses.

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The primary objective of this study was to establish an atmospheric deposition gradient with ecologically analogous sites in order to reduce confounding variables in studies of N deposition effects on forests. Initially we screened 11 sites along a 41-km transect in the SGM in order to select ecologically analogous sites with a range of N deposition inputs. We hypothesized that atmospheric deposition within the SGM varies considerably depending on the air flow patterns produced by the combination of ridge and canyon orientation and the prevailing northeasterly winds (Fig. 1). Based on that premise we hoped to select sites along a N-deposition gradient that had minimal observable environmental variation, other than atmospheric deposition inputs (Kiefer and Fenn, 1997).

Once ecologically analogous sites were selected, N and S deposition were measured in year-round bulk throughfall, and as seasonal dry deposition to paper filters to more fully characterize the deposition gradient (Bytnerowicz et al., 1987a; Fenn and Bytnerowicz, 1993). The N status of pine trees in the plots was also evaluated from tree growth and nutrient responses to N fertilization, and is reported elsewhere (Kiefer and Fenn, 1997).

2. Materials and methods

2.1. Selection of study sites

Three study sites were selected from an initial survey of 11 sites in the mixed-conifer zone in the SGM northeast of Los Angeles, California (Kiefer, 1995). At each of the 11 sites, dry deposition of NO_3^- and NH_4^+ to paper filters (Bytnerowicz et al., 1987a; Fenn and Bytnerowicz, 1993) was measured for two 14-day exposures in late summer 1993. From these 11 sites, three sites (Fig. 1) were chosen for further study based on varying N deposition but otherwise similar site characteristics (soil family, soil texture, elevation, slope, precipitation, aspect, vegetation). At all three sites the soils were of the Lithic Xerorothent family, and site elevation was 2060 m (Kiefer and Fenn, 1997). Deposition presumably differs among the three sites because of varying air flow patterns. The three sites chosen for throughfall collection and seasonal dry deposition measurements, Kratka (KR, relatively high deposition), Vista (VI, relatively moderate deposition) and Rock Creek (RC, relatively low deposition) were no more than 2 km apart. The study areas are located approximately 65 km inland northeast of Santa Monica Bay (Fig. 2).

2.2. Atmospheric deposition measurements

Bulk throughfall samples were collected in the three plots for 1 year. Throughfall was collected using the flip-top rain collectors described by Glaubig and Gomez (1994), except that the collectors were left open (tops were not installed) to facilitate snow collection. Snow was the dominant form of precipitation at these sites. Because the collector openings were continuously open, the samples are considered as bulk deposition. The collectors were composed of a plastic cylinder with a

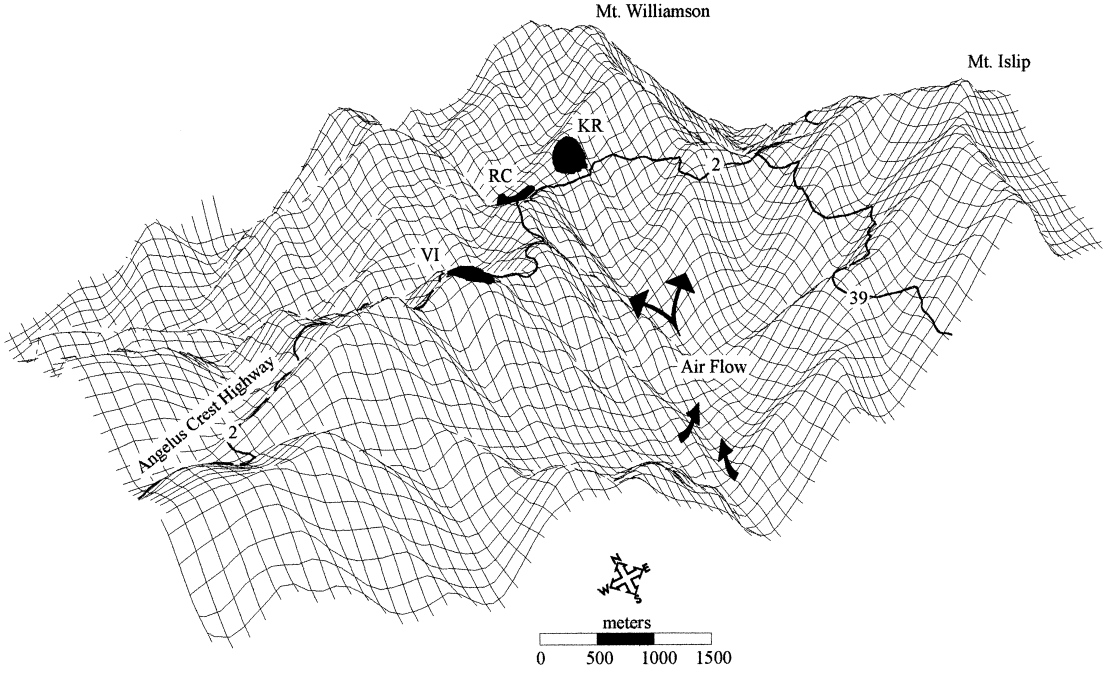

Fig. 1. Three-dimensional mesh plot showing canyon and ridge orientation of the three study sites. The vertical dimension has been exaggerated by a factor of two for easier visualization of the exposure of the three research sites to the prevailing winds. KR, Kratka; RC, Rock Creek; VI, Vista.

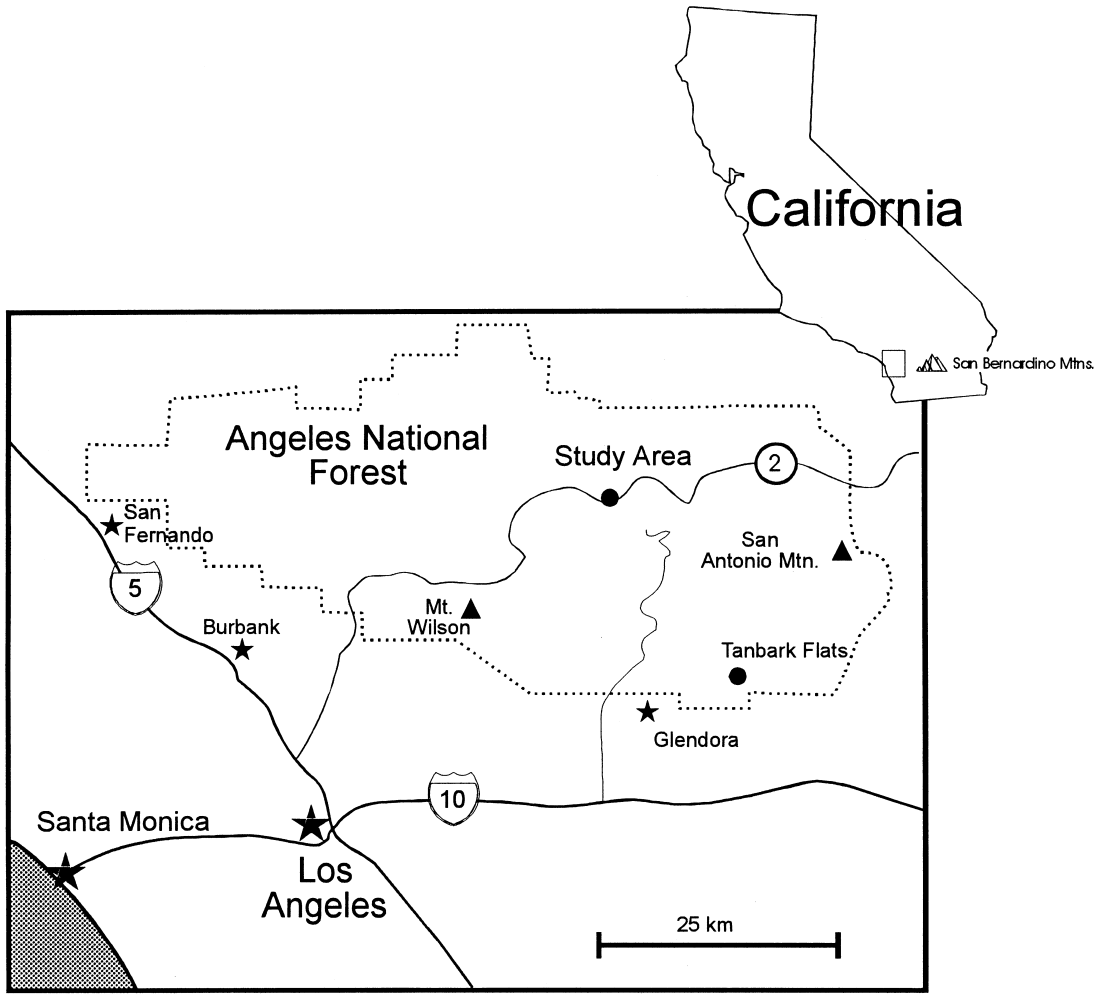

Fig. 2. Location of the study area near Kratka Ridge where throughfall and dry deposition samples were collected. The three plots, Kratka (KR), Vista (VI), and Rock Creek (RC) were no greater than 2 km apart.

diameter of 10 cm, 42 cm in height and a volume of 2.35 liters. The cylinder openings were located approximately 1 m above ground level. The collectors were set in place on 18 December 1993, and the first precipitation collection occurred in January 1994. Fifteen bulk throughfall collectors were placed at each of the three study areas. Twelve of the collectors were placed randomly under tree canopies to sample throughfall and three were placed in the open to collect precipitation. The bulk deposition collectors were scattered over an area of ca. 3 ha within each study site. Throughfall samples were collected immediately after a precipitation event to minimize evaporation and contamination of the samples. Throughfall volumes were measured and an aliquot of each throughfall sample was frozen until analysis for NO_3^-, NH_4^+, and SO_4^{2-}. Ionic concentrations were determined with ion chromatography (Dionex[1] series 4000i, Sunnyvale, CA).

[1] Mention of trade names or products is for information only and does not imply endorsement by the US Department of Agriculture.

The per cent of the study areas covered by overstory canopy was determined using a spherical densiometer (Lemon, 1957). The densiometer consisted of a spherical mirror 15 cm in diameter which was etched with a grid. The densiometer was held so that the mirror reflected the overstory. The per cent canopy cover was determined from the number of quadrants filled with a reflection of the canopy and the number of quadrants not filled. Sampling points for densiometer readings were selected at 20-m intervals with initial points determined randomly. Four readings were taken at each sampling point, with a reading facing north, east, south and west. Forty sampling points were measured in each of the three study areas.

Deposition rates to the forest (kg ha^{-1} $year^{-1}$) were determined as described previously (Fenn and Bytnerowicz, 1997). Bulk throughfall deposition inputs to the forest canopy were calculated by determining the content (concentration×volume) of each ion in the collectors placed under tree canopies. An average value was determined from the 12 collectors under trees per plot.

Deposition per land area covered by canopy (kg ha^{-1} $year^{-1}$) was determined by extrapolation from deposition to the area of the collector openings (79 cm^2 per collector) to the area of the plot covered with overstory. This same procedure was done for the collectors placed in open areas to determine annual deposition in bulk precipitation. Combining annual totals for precipitation deposition in the open areas with gross throughfall deposition under the canopy areas provided an estimate of total bulk throughfall deposition in the plot. Net throughfall, a measure of dry deposition, was calculated as total deposition in the plot minus deposition in precipitation for the entire plot.

Dry deposition of NH_4^+, NO_3^-, and SO_4^{2-} to surrogate surfaces at the three study sites was also determined during the summer dry season as previously described (Bytnerowicz et al., 1987a; Fenn and Bytnerowicz, 1993). Whatman no. 41 paper filter disks, 47 mm in diameter were placed in polycarbonate filter holders oriented horizontally 2 m above ground level in canopy-free areas. Eight filter disks were randomly located per study area. Filters were exposed for 2-week periods without rain from 24 June 1994 until 29 September 1994. After the 2-week exposure the disks were extracted in deionized distilled water and ionic content of the filter extracts was determined by ion chromatography. Concentrations in the extracts were corrected for ions extracted from unexposed filter blanks. The exposed surface areas of the upper and lower sides of the filters were used to calculate deposition fluxes.

Data analysis was performed using SigmaStat™ statistical software from Jandel Scientific Software (San Rafael, CA). Differences between concentrations of NO_3^-, NH_4^+, and SO_4^{2-} in precipitation and in throughfall were tested by one-way analysis of variance (ANOVA) and Tukey's all pairwise multiple comparison procedures. Comparisons of dry deposition fluxes to surrogate surfaces were tested by repeated measures one-way ANOVA and Tukey's multiple comparison test. Differences were considered statistically significant at $p < 0.05$ (Fox et al., 1994).

3. Results

Total inorganic N deposition (NH_4^+ plus NO_3^-) was 6.0, 8.6 and 11.5 kg ha^{-1} $year^{-1}$ at RC, VI and KR. Total SO_4-S deposition was 7.8, 12.5 and 9.7 kg ha^{-1} $year^{-1}$ at RC, VI and KR (Fig. 3). Nitrate deposition in net throughfall was much greater than in precipitation at all three sites (Fig. 4). Likewise, NH_4^+ deposition was greater in net throughfall than in precipitation at RC and KR. At VI, however, NH_4^+ deposition in net throughfall was only 30% as great as NH_4^+ deposition in precipitation. Sulfate deposition was relatively low in net throughfall compared to precipitation at all three

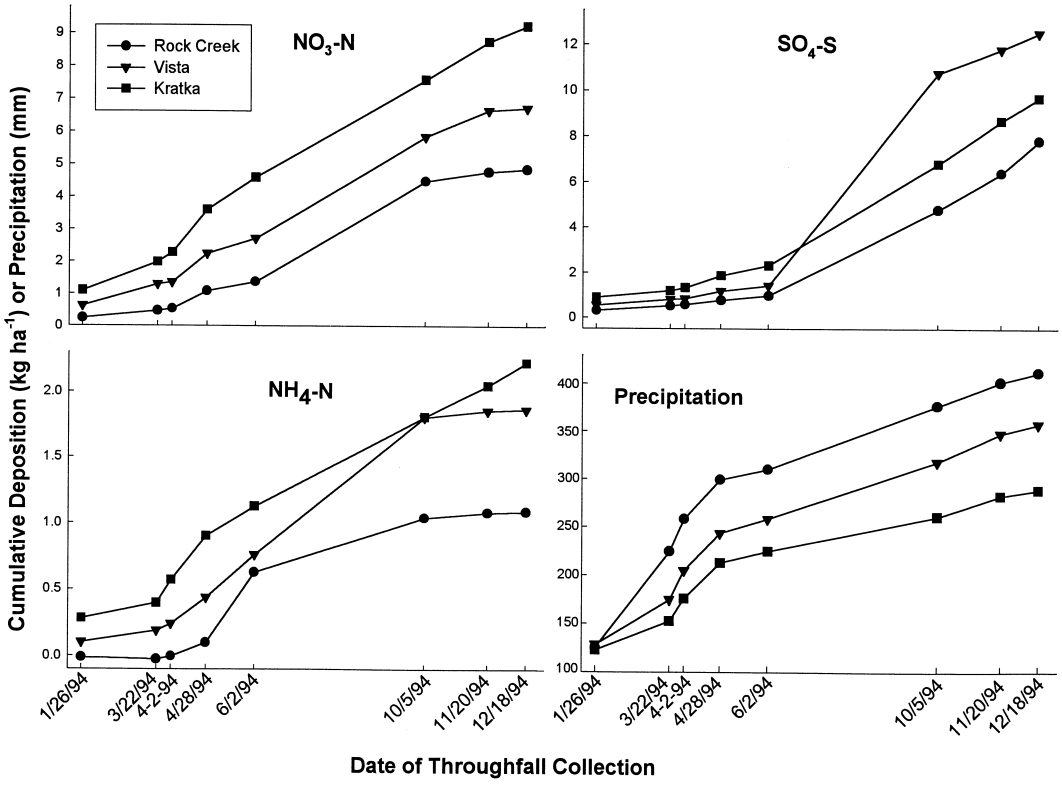

Fig. 3. Cumulative bulk throughfall deposition and precipitation in 1994 at three sites in the San Gabriel Mountains.

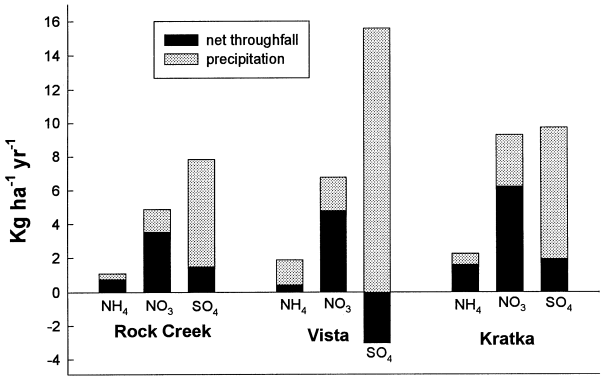

Fig. 4. Net throughfall (total throughfall deposition minus precipitation deposition) and deposition in precipitation (mainly as snowfall) in 1994. Deposition values are given as NH_4-N, NO_3-N, and SO_4-S.

sites. In fact, at VI, estimated SO_4^{2-} deposition in net throughfall was a negative value (-3.0 kg S ha^{-1} $year^{-1}$), because average deposition in the collectors in the open areas was greater than that collected in the throughfall collectors (Fig. 4). Greater SO_4^{2-} in the open collectors suggests that atmospheric SO_4^{2-} may have been partially retained by the canopy. Total annual precipitation to the open collectors at RC, VI and KR was 412, 358 and 289 mm, respectively (Fig. 3).

Volume-weighted mean ion concentrations of NO_3^- in precipitation and throughfall were generally greater than those of NH_4^+ at RC and KR. Ammonium concentrations were significantly lower than SO_4^{2-} concentrations in precipitation and throughfall in the three sites in nearly every case (Fig. 5). However, at VI concentrations of SO_4^{2-} (274 μeq $liter^{-1}$; SE 25) in precipitation were unusually high compared to NO_3^- concentrations (39 μeq $liter^{-1}$; SE 7). Sulfate concentrations in throughfall were similar to NO_3^- concentrations at all three study sites (Fig. 5).

Deposition fluxes of NO_3^- and NH_4^+ to paper filters demonstrated increasing dry deposition from RC to VI, to KR. Graphical presentation of these data are essentially parallel lines showing clear separation between N deposition fluxes in the three study sites (Fig. 6). However, dry deposition of SO_4^{2-} was virtually identical at the three sites, and was an order of magnitude lower than dry deposition of NO_3^- and NH_4^+. Dry deposition fluxes of NO_3^- to the filters was slightly greater than deposition of NH_4^+. Dry deposition of NO_3^- and NH_4^+ peaked in mid-August and then declined to a constant level for the rest of the summer. In contrast, dry deposition of SO_4^{2-} was at its highest in the latter part of the summer (Fig. 6).

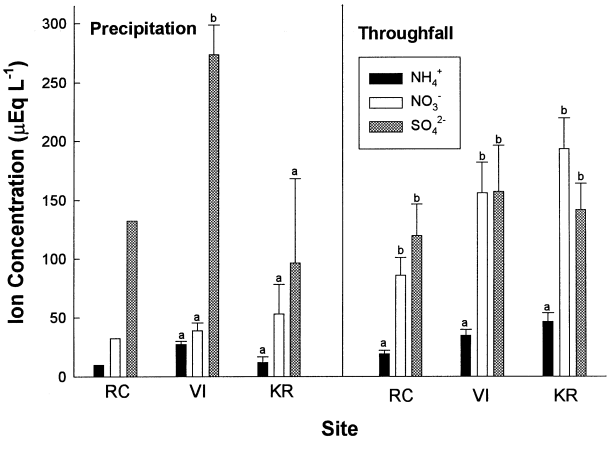

Fig. 5. Volume-weighted annual mean ion concentrations in bulk precipitation and bulk throughfall. Letters above bars indicate significant differences between ions at each site. Error bars represent standard errors of the site means. No statistical tests were performed for the precipitation data at Rock Creek (RC) because on some dates precipitation samples were collected from only one collector located in the open areas. VI, Vista; KR, Kratka.

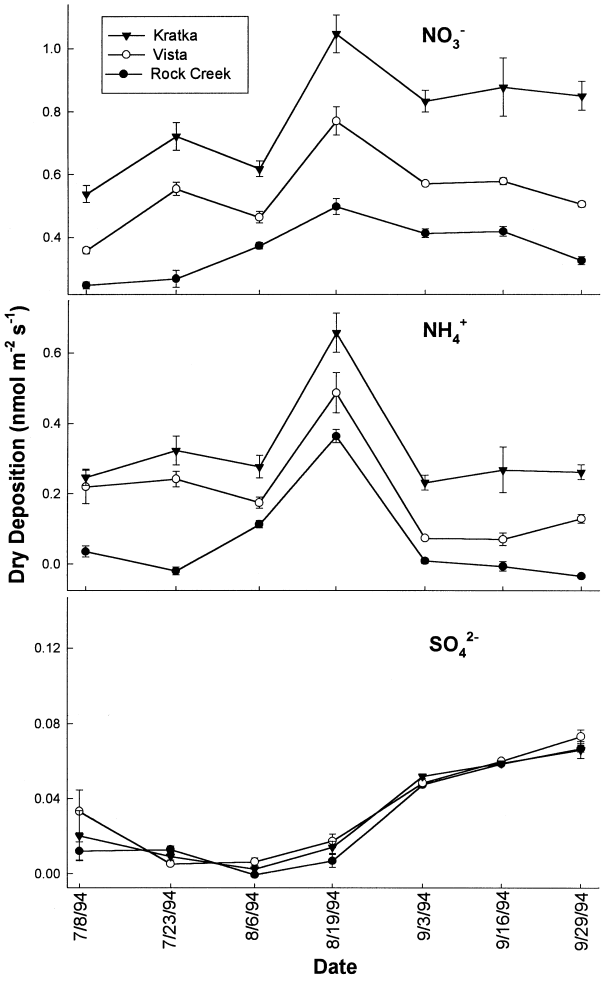

Fig. 6. Average dry deposition fluxes to paper filter disks at the three sites during the summer dry season. Dry deposition fluxes of NO_3^- and of NH_4^+ were significantly different among the three sites ($p < 0.05$). Deposition fluxes of SO_4^{2-} were not significantly different among the three sites.

4. Discussion

4.1. N and S deposition

Studies of air pollution inputs to forests and shrub lands in southern California have focused on ozone (Miller et al., 1994) and dry deposition of nitrogenous pollutants (Bytnerowicz and Fenn, 1996) because of the severity of smog episodes during the long dry summers and because N emissions are much greater than S emissions in the south coast (Los Angeles) air basin. Emissions of SO_x in the south coast air basin in 1990 were less than 10% as high as NO_x emissions (South Coast Air Quality Management District, 1994). Major anthropogenic S sources are primarily limited to oil refineries in the Long Beach area, near the coast south of Los Angeles. However, in our study S deposition in bulk throughfall (7.8–12.5 kg ha^{-1} $year^{-1}$) was of similar magnitude to N deposition (6.0–11.5 kg ha^{-1} $year^{-1}$). The high S inputs were unexpected because of relatively low levels of S emissions and previous reports of low S deposition (Bytnerowicz et al., 1987a, b; Fenn and Bytnerowicz, 1993). Moderately elevated N deposition was expected based on high levels of atmospheric N pollutants in low-elevation chaparral watersheds in the SGM (Riggan et al., 1985; Bytnerowicz et al., 1987a, b; Grosjean and Bytnerowicz, 1993), in the mixed-conifer zone in the San Bernardino Mountains (Fenn and Bytnerowicz, 1993, 1997), and in regional studies (Solomon et al., 1992; Bytnerowicz and Fenn, 1996). On the other hand, throughfall and precipitation inputs of S as high or higher than N at our sites is supported in some instances by data on precipitation inputs in the SGM. Waldman et al. (1985) reported that SO_4^{2-} concentrations in wet deposition were similar to NO_3^- concentrations at Heninger Flats (780 m), a pine forest located on the southern slope of Mount Wilson in the SGM. At Mount Wilson (1736 m), which is 20-km southwest of KR, volume-weighted mean concentrations of SO_4^{2-} in precipitation were more than twice as high as NO_3^- concentrations over a 1-year period (Liljestrand and Morgan, 1981). Data from the California Acid Deposition Monitoring Program for 1985–90 (Blanchard and Tonnessen, 1993) show a 1.15 NO_3:SO_4 equivalent ratio in precipitation at Mount Wilson and a 1.47 ratio at Tanbark Flats (817 m), a chaparral site in the San Dimas Experimental Forest. Tanbark Flats is 920 m lower in elevation than Mount Wilson, is more directly exposed to N-polluted air masses from the Los Angeles metropolitan area, and is located ca. 28 km further inland than Mount Wilson.

Nitrate deposition in throughfall and to filter disks was highest at KR, intermediate at VI and lowest at RC. The primary sources of atmospheric NO_3^- are fossil fuel emissions in the Los Angeles urban area (Solomon et al., 1992). Dry-deposited N compounds accumulate on canopy surfaces, resulting in washoff of NO_3^- during precipitation events. A lesser amount of nitrogenous pollutants are washed directly out of the atmosphere by precipitation. We attribute differences in bulk throughfall NO_3^- deposition and in dry deposition fluxes to surrogate surfaces among the sites to varying exposures to incoming N-bearing air masses (Fig. 1). Deposition of NH_4^+ was also attributed to dry deposition of NH_x and washoff of NH_4^+ from canopy surfaces. NH_4^+ concentrations and deposition in throughfall and to surrogate surfaces at RC and KR were lower than for NO_3^- as previously documented for the Los Angeles air basin (Bytnerowicz et al., 1987a; Bytnerowicz and Fenn, 1996). However, at Camp Paivika in the western San Bernardino Mountains, NH_4^+ deposition in throughfall and fog is similar to NO_3^- deposition because of the high concentration of large dairy operations in the Chino area west of Camp Paivika (Fenn and Bytnerowicz, 1997).

4.2. Marine influence on atmospheric deposition inputs

At our sites in the high-elevation mixed-conifer forest, the NO_3:SO_4 ratio in bulk precipitation ranged from 0.14 to 0.44, and 0.84 to 1.30 in bulk throughfall. These values are not directly comparable to wet-only deposition values discussed above, because of the dry deposition component in bulk deposition. Nonetheless, the ratios for RC, VI and KR indicate the greater magnitude of SO_4^{2-} deposition in these sites compared to sites with greater N deposition and greater distance from the ocean (e.g. Tanbark Flats and San Bernardino Mountain sites). The low NO_3:SO_4 ratios in bulk deposition at our sites suggest that marine aerosols influence atmospheric inputs there, because no other major sources of S are known in this area. Furthermore, anthropogenic S sources would be expected to increase dry deposition of SO_4^{2-}, but at our sites dry deposition of SO_4^{2-} was minimal.

All of the storms that occurred during this study originated over the Pacific Ocean, suggesting the potential for sea salt inputs of SO_4^{2-} at sites within the zone of coastal influence. Wet deposition data from Tanbark Flats support our conclusion that S deposition at our study sites was mainly of marine origin. For example, a large increase in throughfall S deposition was measured in our sites in early October 1994. Wet deposition data during this same storm showed a corresponding large influx of Na^+, Cl^- and SO_4^{2-} at Tanbark Flats (NADP, 1994). The Cl^-:Na^+ ratio in precipitation samples can be used to determine the contribution of sea salt to deposition inputs. Likewise, SO_4^{2-}:Na^+ ratios can be used to determine the importance of marine sources of SO_4^{2-} (Millet et al., 1997). For seawater the Cl^-:Na^+ ratio is ≈ 1.17 and the SO_4^{2-}:Na^+ ratio is 0.12 (ions expressed as μeq $liter^{-1}$). Ratios for precipitation

collected at Tanbark for the week of 4–11 October 1994 were 1.14 for Cl^-/Na^+ and 0.35 for SO_4^{2-}:Na^+. These ratios suggest a strong marine influence, although not all of the SO_4^{2-} deposited at Tanbark can be attributed to sea salt deposition.

Precipitation and throughfall are generally influenced by sea salt at sites within approximately 100 km from the coastline (Parker, 1983). Our sites are 65 km inland, which suggests that a moderate marine influence on atmospheric deposition can be expected. The prevailing air flow pattern in the Los Angeles basin is on-shore from southwest to northeast. The onshore flow patterns undoubtedly have the potential to transport sea salt components inland. Unfortunately, we cannot definitively evaluate the strength of the marine influence on deposition at our sites, since the precipitation and throughfall samples were not analyzed for Na^+ or Cl^-. Collecting throughfall and precipitation samples along a transect from the coast to sites further inland and analyzing samples for major ions as reported for coastal Washington state (Blew and Edmonds, 1995) would provide more definitive information on the relative importance of sea salt inputs. In a previous transect study in California, extending from a marine site (Santa Cruz Island) to a coastal site (Santa Monica Mountains), and to two inland sites (Tanbark Flats in the SGM, and Sequoia National Park in the southwestern Sierra Nevada Mountains) it was demonstrated that SO_4^{2-} concentrations in bulk deposition decreased with greater distance from the ocean (Paul Miller, unpublished data). The source of increased NH_4^+ deposition at VI in October is not known, but may be a result of marine aerosols aged by contact with land-based emissions. Aged marine aerosols are known to have higher concentrations of secondary ammonium sulfate and nitrate (Chow et al., 1996).

4.3. Wet and dry deposition

Our data clearly show that SO_4^{2-} deposition was predominantly in wet form at all three study sites. This was especially true at VI where the estimated SO_4^{2-} deposition in precipitation was so high that a negative value for SO_4^{2-} deposition in net throughfall was obtained. Storm transport and wet deposition of sea salt SO_4^{2-} is likely the main cause of high wet deposition inputs of SO_4^{2-} in this study. Additional contributing factors could include the lower dry deposition rates of S pollutants compared to N pollutants (Young et al., 1988), and less effective precipitation scavenging of N oxides compared to other compounds such as sulfur dioxide (Liljestrand and Morgan, 1981). By comparison, NH_4^+ deposition at RC and KR, and NO_3^- deposition at all three sites were much greater in net throughfall than in precipitation, presumably because of the greater importance of dry deposition of these nitrogenous pollutants. Calculations of annual dry deposition of N (kg ha^{-1} $year^{-1}$) at our sites using the surrogate surface deposition flux data gave estimates which agreed well with the net throughfall data. However, only crude comparisons can be made, since dry deposition data to the filters are only available for the summer dry season.

4.4. Nitrogen and S deposition, N saturation, and foliar S

N fertilization experiments clearly demonstrated that notwithstanding the moderate throughfall deposition inputs at our sites (6.0–11.5 kg N ha^{-1} $year^{-1}$ in bulk throughfall), Jeffrey pine trees in the plots are still N-limited (Kiefer and Fenn, 1997). By comparison, ponderosa pine (*Pinus ponderosa* Laws.) trees in the western San Bernardino Mountains (Camp Paivika) where N deposition inputs in throughfall were $\geqslant$19 kg ha^{-1} $year^{-1}$ (Fenn, unpublished data) did not respond to N fertilization (Kiefer and Fenn, 1997), and a number of plant, soil and hydrologic parameters indicated that the forest there is N saturated (Fenn et al., 1996). Chaparral watersheds in the San Dimas Experimental Forest (includes Tanbark Flats; throughfall N deposition 23 kg ha^{-1} $year^{-1}$) in the SGM are also N saturated based on elevated export of NO_3^- in streamwater (Riggan et al., 1985, 1994) and high NO emissions from soil (Anderson et al., 1988). Throughfall deposition of N frequently underestimates total deposition by 25–50% (Miller et al., 1993; Lovett and Lindberg, 1993; Garten et al., 1998), especially when NH_4^+ is a significant component of the total N deposition such as at Camp Paivika in the western San Bernardino Mountains (Fenn and Bytnerowicz, 1997). Based on throughfall data from Tanbark Flats and Camp Paivika, total N deposition in these N-saturated sites is estimated to be 25–35 kg ha^{-1} $year^{-1}$ compared to 9–16 kg ha^{-1} $year^{-1}$ in the three N-limited sites in the upper San Gabriel Mountains. In general, these Mediterranean-climate ecosystems in southern California and European forests seem to have similar N deposition thresholds at which symptoms of N saturation become apparent. In a survey of 65 European forests, N leaching was observed in all forests with N deposition greater than 25 kg ha^{-1} $year^{-1}$, only at some sites with intermediate N deposition levels (10–25 kg ha^{-1} $year^{-1}$), and no significant leaching occurred at sites with N deposition less than 10 kg ha^{-1} $year^{-1}$ (Dise and Wright, 1995).

Elevated S deposition in the three SGM sites is reflected in foliar S content. Foliar concentrations of S at RC, VI, and KR were 0.70, 0.69, and 0.83 g kg^{-1}, and were significantly higher ($p < 0.05$) than foliar S concentrations at four sites in the San Bernardino Mountains (0.48–0.56 g kg^{-1}). Sulfur deposition across an air pollution gradient in the San Bernardino Mountains was estimated to range from 0.9–2.9 kg ha^{-1} $year^{-1}$ (Fenn and Bytnerowicz, 1993). These data support the hypothesis that elevated wet deposition of SO_4^{2-} is an

ongoing chronic phenomenon at the three SGM sites and contributes to higher plant uptake and accumulation of S (Rennenberg, 1984).

5. Conclusions

Although the three study sites were only 2 km apart, dry deposition of NO_3^- and NH_4^+ to surrogate surfaces during the dry 'smog' season and total annual bulk throughfall deposition of N were consistently highest at KR, intermediate at VI, and lowest at RC. Bulk N and S deposition were of similar magnitude in each of the three plots, contrary to more inland montane sites where wet and dry deposition of S is typically several-fold lower than N deposition. Higher SO_4^{2-} deposition in bulk precipitation and throughfall in this study compared to montane sites further inland was presumably because of marine aerosols transported inland during storm events and by the prevailing winds. The location of the study sites in relation to the ocean (65 km inland) and to the urban smog source, resulted in throughfall inputs which were higher in S (7.8–12.5 kg ha^{-1} $year^{-1}$), but lower in N (6.0–11.5 kg ha^{-1} $year^{-1}$) than montane sites in the lower SGM (23 kg N ha^{-1} $year^{-1}$) or in the western San Bernardino Mountains further inland (19 kg N ha^{-1} $year^{-1}$; 3 kg S ha^{-1} $year^{-1}$). N fertilization experiments at our sites clearly demonstrated that despite moderate N deposition inputs, Jeffrey pine trees in the plots are highly N-limited (Kiefer and Fenn, 1997). However, elevated wet deposition of S in the three study sites appears to contribute to increased S concentrations in Jeffrey pine foliage compared to montane sites in the Los Angeles air basin with low S deposition. The results of this and other studies suggest that N saturation symptoms such as lack of tree growth response to N fertilization, elevated nitric oxide emissions from soil, or elevated NO_3^- export in streamwater do not occur in southern California montane ecosystems until throughfall levels reach a threshold of approximately 16–19 kg ha^{-1} $year^{-1}$.

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