Throughfall and fog deposition of nitrogen and sulfur at an N-limited and N-saturated site in the San Bernardino Mountains, southern California

Mark E. Fenn, Mark A. Poth, Susan L. Schilling, and David B. Grainger

Abstract: Inorganic nitrogen (N) and sulfur (S) deposition in bulk throughfall and fog were determined at two sites located at opposite ends (42 km apart) of a pollution gradient in the San Bernardino Mountains. Plot-level averages for total annual N and S deposition in throughfall in 1996 were 18.8 and 2.9 kg·ha^{-1}, respectively, at Camp Paivika (CP) and 2.9 and 0.4 kg·ha^{-1}, respectively, at Barton Flats (BF). Deposition of N in throughfall in the four transects at CP ranged from 12.1 to 31.7 kg·ha^{-1}·year^{-1}. Spatial variability was high because of heterogenous canopy cover and varying exposure to air pollution. Annual estimated stand-level deposition of N and S in fog were 10.9 and 1.9 kg·ha^{-1}, respectively, at CP, and 0.6 and 0.2 kg·ha^{-1}, respectively, at BF. We estimated that N deposition in fog contributed 35% of the total annual N deposition at CP and 13% at BF. Analogous values for S were 39% at CP and 26% at BF. Ammonium deposition in throughfall and fog at CP were 72 and 122% as high as NO_3^- deposition because of elevated NH_3 emissions from dairy farms in the Chino–Norco area 34 km southwest of CP.

Résumé : Les dépôts de soufre (S) et d'azote (N) inorganiques contenus dans le pluviolessivat et la brume ont été mesurés sur deux sites distants de 42 km et localisés aux deux extrémités d'un gradient de pollution dans les montagnes de San Bernardino. Les moyennes des parcelles pour le dépôt annuel total de N et de S dans le pluviolessivat en 1996 étaient 18,8 et 2,9 kg·ha^{-1} à Camp Paivika (CP) et 2,9 et 0,4 kg·ha^{-1} à Barton Flats (BF). Le dépôt de N dans le pluviolessivat des quatre transects à CP variait de 12,1 à 31,7 kg·ha^{-1}·a^{-1}. La variation spatiale était élevée à cause de l'hétérogénéité du couvert et de l'exposition variable à la pollution de l'air. À l'échelle du peuplement, le dépôt annuel de N et de S dans la brume était respectivement de 10,9 et 1,9 kg·ha^{-1} à CP et de 0,6 et 0,2 kg·ha^{-1} à BF. Nous estimons que le dépôt de N dans la brume représente 35% du dépôt annuel total de N à CP et 13% à BF. Les valeurs analogues pour S sont de 39% à CP et de 26% à BF. Le dépôt d'ammonium dans le pluviolessivat et la brume à CP représente 72 et 122% du dépôt de NO_3^- à cause des fortes émissions de NH_3 provenant des fermes laitières dans la région de Chino/Norco à 34 km au sud-ouest de CP.

[Traduit par la Rédaction]

Introduction

Human alteration of the nitrogen (N) cycle is an environmental and ecological issue of major concern, over spatial scales ranging from a forest stand to global (Vitousek et al. 1997). A growing number of studies have identified forested areas exposed to chronic atmospheric N deposition that exhibit symptoms of N excess, analogous to overfertilization of arable land (Fenn et al. 1998; Stoddard 1994). Increased ecosystem N loss is a symptom of N overload and results in degradation of water quality and potentially deleterious effects on aquatic systems that are recipients of N runoff from terrestrial watersheds (Fenn et al. 1998; Stoddard 1994). Degraded water quality (e.g., elevated NO_3^-) in forested catchments can be of particular concern when these water supplies are relied upon for drinking water (Lovett et al. 2000; Murdoch and Stoddard 1992) or as pristine sources to improve the quality of polluted water sources (Riggan et al. 1985).

Nitrogen deposition rates in the more exposed regions of the San Bernardino and San Gabriel Mountains near Los Angeles, Calif., are among the highest in North America (Bytnerowicz and Fenn 1996). Anthropogenic S deposition, however, is typically 7–10 times lower than N deposition (Bytnerowicz et al. 1987*a*; Fenn and Bytnerowicz 1993). Widespread reliance on fossil fuels for transportation and energy production is the primary source of atmospheric N oxides (Alexis et al. 1999). The elevated load of N oxides emitted to the atmosphere in urban areas leads to chronic N deposition in montane ecosystems that border the South Coast (Los Angeles) Air Basin (SCAB). The N oxides also function as precursors in the photochemical reactions of ozone formation. Nitrogen deposition in the SCAB is dominated by dry deposition of oxidized forms of N except in areas downwind of the concentrated dairy operations in the Chino–Norco area, which provide a source of reduced N (NH_x) pollutants (Russell and Cass 1986). Nitrogen deposition in fog is also important in montane areas with high air pollution exposure and frequent fog occurrence (Collett et al. 1989; Miller et al. 1993).

Chaparral ecosystems and mixed conifer forests in the SCAB with elevated N deposition exhibit the classic symp-

Received December 20, 1999. Accepted June 14, 2000.

M.E. Fenn,[1] M.A. Poth, S.L. Schilling, and D.B. Grainger. USDA Forest Service, Pacific Southwest Research Station, 4955 Canyon Crest Drive, Riverside, CA 92507, U.S.A.

[1]Corresponding author. e-mail: mfenn@deltanet.com

© 2000 NRC Canada

toms of N saturation, such as elevated NO_3^- concentrations in soil solution, high leachate losses of NO_3^- and cations, high export of NO_3^- in stream water (Fenn and Poth 1999), elevated emissions of nitrogenous trace gases from soil, and high N and N/nutrient ratios in foliage and litter (Fenn et al. 1996). Chronic N deposition, in conjunction with high ozone exposure and long-term fire suppression in these systems, constitute major ecological and environmental perturbations. Impacted water quality due to high nitrate concentrations in streams and groundwater from montane watersheds is of particular concern considering the high value of water in the highly populated semiarid region surrounding Los Angeles and adjacent counties. Nitrate levels in stream water from polluted chaparral and forested watersheds in the SCAB are the highest values reported for wildland watersheds in North America (Stoddard 1994; Fenn and Poth 1999; Riggan et al. 1994; Riggan et al. 1985).

Nitrogen enrichment, as a result of chronic N deposition, also causes significant effects on the forest ecosystem (Takemoto et al. 2000). Recent studies in the western high-deposition areas of the San Bernardino Mountains (SBM) show that net nitrification rates are high with rapid and complete nitrification of available ammonium (M.E. Fenn and M.A. Poth, unpublished data). This high nitrification activity is a key process leading to elevated N loss from the system as leached nitrate and as gaseous nitric oxide (NO) emissions from soil (Fenn et al. 1996). Standing fine-root biomass and belowground allocation of carbon in ponderosa pine trees in the high-deposition sites in the SBM has been dramatically reduced, presumably because of ozone stress and high N fertility (Grulke et al. 1998). The phenology of ponderosa pine (*Pinus ponderosa* Laws.) trees at Camp Paivika, in the western SBM, has also been dramatically altered with 95% of total foliar biomass found in the current-year foliage. This phenological behaviour, as a result of high N deposition and ozone exposure, has been likened to that of deciduous species (Grulke and Balduman 1999). The combined effects of ozone and nitrogen have also resulted in unusually high litter accumulation in the forest floor as a result of greater foliage production and accelerated litter fall, a response which was also predicted by the Century simulation model (Arbaugh et al. 1999).

Year-round deposition inputs to natural ecosystems in the SCAB have only been reported in two throughfall studies in the San Gabriel Mountains (Fenn and Kiefer 1999; Riggan et al. 1985). Studies reporting ionic fog-water deposition or dry deposition in montane areas of southern California report seasonal data only (Waldman et al. 1985; Fenn and Bytnerowicz 1997; Bytnerowicz et al. 1987*a*). Notwithstanding the important consequences of N deposition and the long history of air pollution research in the SBM, deposition inputs of N and S at the forest stand level have not been adequately determined. This is largely due to the difficulty in quantifying fog and dry deposition, particulary in heterogeneous terrain (Lovett 1994; Lovett et al. 1997). Most of the air pollution studies in the SCAB have focused on urban areas, on atmospheric pollutant concentrations, or dry deposition fluxes to branches or surrogate surfaces (Bytnerowicz et al. 1987*a*; Fenn and Bytnerowicz 1993). Fenn and Bytnerowicz (1997) reported throughfall deposition for 3 years at Barton Flats, a relatively low N deposition site in the SBM, but throughfall sampling was not done during winter, which is the dominant wet season in this Mediterranean climate.

Because of the high cost and difficulty of measuring dry deposition fluxes in forest stands, quantifying N and S deposition in throughfall provides a useful alternative measure of annual atmospheric inputs at the stand level. Throughfall deposition provides a reliable estimate of total S deposition to a forest (Butler and Likens 1995; Lovett 1994). However, in areas of chronic N deposition, throughfall frequently underestimates total (wet and dry) N deposition (Fenn and Bytnerowicz 1997; Lovett and Lindberg 1993). Underestimates of deposition by the throughfall method are attributed to canopy uptake and retention of atmospheric N compounds, especially from dry deposition (Garten et al. 1998). Notwithstanding this limitation, throughfall estimates of N deposition are useful in setting the lower bounds of N deposition. Because of the large worldwide database of throughfall deposition to forests (Lovett and Lindberg 1993; Parker 1983; Thimonier 1998), throughfall inputs can be used to compare with N inputs in other areas, thus providing an indication of the relative severity of N deposition.

As described above, studies of various aspects of atmospheric deposition in the SBM have been reported. However, none of the studies reports year-round deposition inputs in wet and dry forms across the deposition gradient. Therefore, the major objective of this study was to quantify landscape-level (per hectare) annual throughfall deposition of N and S at a western (Camp Paivika) and eastern (Barton Flats) site in the SBM and to estimate the importance of fog deposition. Data on atmospheric deposition inputs are needed to better delineate the atmospheric deposition loadings at which ecological effects of added N are observed in these western forests.

Materials and methods

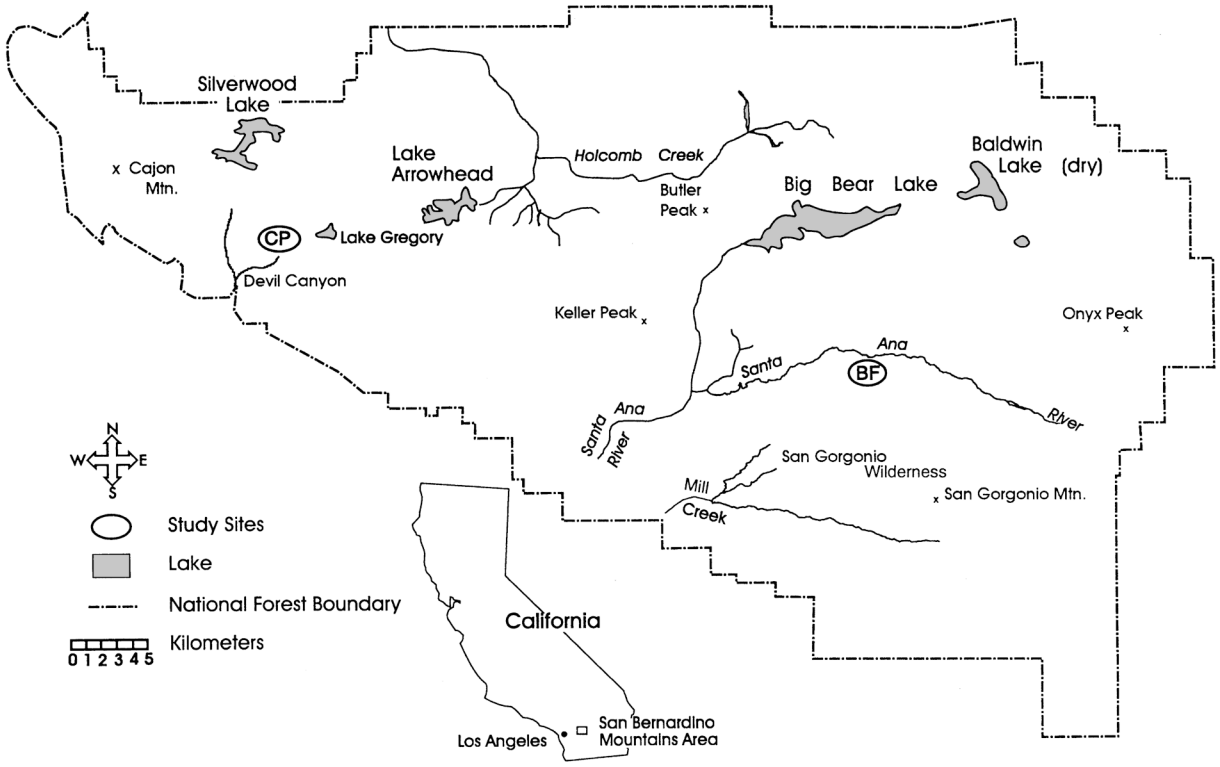
Study sites

Throughfall and fog-water samples were collected at Camp Paivika (CP) and Barton Flats (BF), two mixed-conifer forest sites located at opposite ends of the west–east air pollution gradient in the SBM (Fig. 1; Fenn and Bytnerowicz 1993). Camp Paivika (1580 m elevation) is a high pollution site located at the extreme western end of the mixed conifer zone in the SBM. Ozone levels in the adjacent mountain community of Crestline are now the highest reported for the SCAB (Alexis et al. 1999). Average annual precipitation at a monitoring station 3.4 km east of CP is 987 mm, and the yearly mean temperature is 12.9°C (Fenn and Poth 1999). Major overstory species at CP are ponderosa pine, incense cedar (*Calocedrus decurrens* (Torr.) Florin.), California black oak (*Quercus kelloggi* Newb.), and to a lesser degree, sugar pine (*Pinus lambertiana* Dougl.). The dense understory at CP is dominated by bracken fern (*Pteridium aquilinum* var. *pubescens* Underw.).

Barton Flats (1945 m elevation) is 42 km ESE of Camp Paivika (Fig. 1). Barton Flats is exposed to moderate ozone concentrations compared with CP and relatively low N deposition based on previous dry deposition measurements (Bytnerowicz et al. 1999*b*). Average annual precipitation is 608 mm, and the mean annual temperature is 10.6°C (Fenn and Poth 1999). The dominant overstory species at BF are ponderosa pine and the closely related Jeffrey pine (*Pinus jeffreyi* Grev. & Balf.), white fir (*Abies concolor* Gord. & Glend.) and California black oak. The sparse

© 2000 NRC Canada

Fig. 1. Map showing location of study sites in the San Bernardino Mountains. CP, Camp Paivika; BF, Barton Flats.



understory at BF is composed of a few shrubs and assorted herbaceous species.

Throughfall collection

Bulk throughfall was collected at CP and BF from January 23, 1996 to January 8, 1997. Throughfall was collected in cylindrical (10 cm diameter) rain gauges. Throughfall was collected on an event basis. When no precipitation occurred for several weeks the collectors were replaced with clean cylinders prior to the next precipitation event to reduce contamination from debris and dry deposition. Collectors were placed in four parallel transects at each site with eight collectors per transect, for a total of 32 collectors per site. Along each transect, collectors were placed 8 m apart. The throughfall collectors at BF were placed in the Barton Flats plot 2 study site, one of three experimental plots established in 1991 as part of a multidisciplinary study of atmospheric deposition effects on the mixed conifer forest (Fenn and Bytnerowicz 1997). At BF the transects were 20 m apart. At CP one pair of north–south transects (20 m apart) was located on a west-facing slope, and the second pair of north–south transects were located on the adjacent east-facing slope (ca. 50 m west of the first pair of transects).

To evaluate factors contributing to the variability in throughfall among the transects within a site, leaf area index (LAI) of each of the four transects at each site was estimated with an indirect optical method (Pierce and Running 1988). Canopy transmittance was measured in April 1998 using an integrating radiometer (Ceptometer, model SF-80, Decagon Devices, Inc., Pullman, Wash.).[2] Radiation measurements were taken in circular "sweeps" consisting of 16 measurements at each point where a throughfall collector was placed. Because the radiometer consists of 80 sensors, this resulted in 1280 point measurements of photosynthetically active radiation (PAR) per throughfall collector location. The data from each of the eight points in a transect were saved in memory and used to calculate an LAI value for each transect.

Throughfall volumes were determined by weight. An equal percent volume was collected from each of the eight collectors per transect and composited for chemical analysis. Ammonium (NH_4^+), nitrate (NO_3^-), and sulfate (SO_4^{2-}) concentrations were determined in the throughfall samples using liquid ion chromatography (Dionex series 4000i, Sunnyvale, Calif.). Throughfall deposition ($kg \cdot ha^{-1} \cdot year^{-1}$) was determined by multiplying the volume of water collected by ionic concentration divided by the area of the collector openings. Deposition at each site (CP and BF) was determined as the average of the four replicate transects. Net throughfall was calculated as total throughfall deposition in the plot minus wet deposition collected in a NADP wet–dry bucket collector located in an open area. Wet deposition data for BF is from the BF monitoring station (Fenn and Bytnerowicz 1997), and wet deposition data for CP is estimated from the National Atmospheric Deposition Program/National Trends Network data from Tanbark Flat, a high deposition site in the San Gabriel Mountains, also in the SCAB, located 55 km west of CP (Bytnerowicz et al. 1987*b*). Tanbark Flat is 750 m lower in elevation than CP but is also highly exposed to urban smog. However, differences in wet deposition are not expected to be great between Tanbark and CP based on previous dry deposition data at the two sites (Bytnerowicz et al. 1987*b*; Fenn and Bytnerowicz 1993) and because wet deposition is minor compared with dry deposition at these sites.

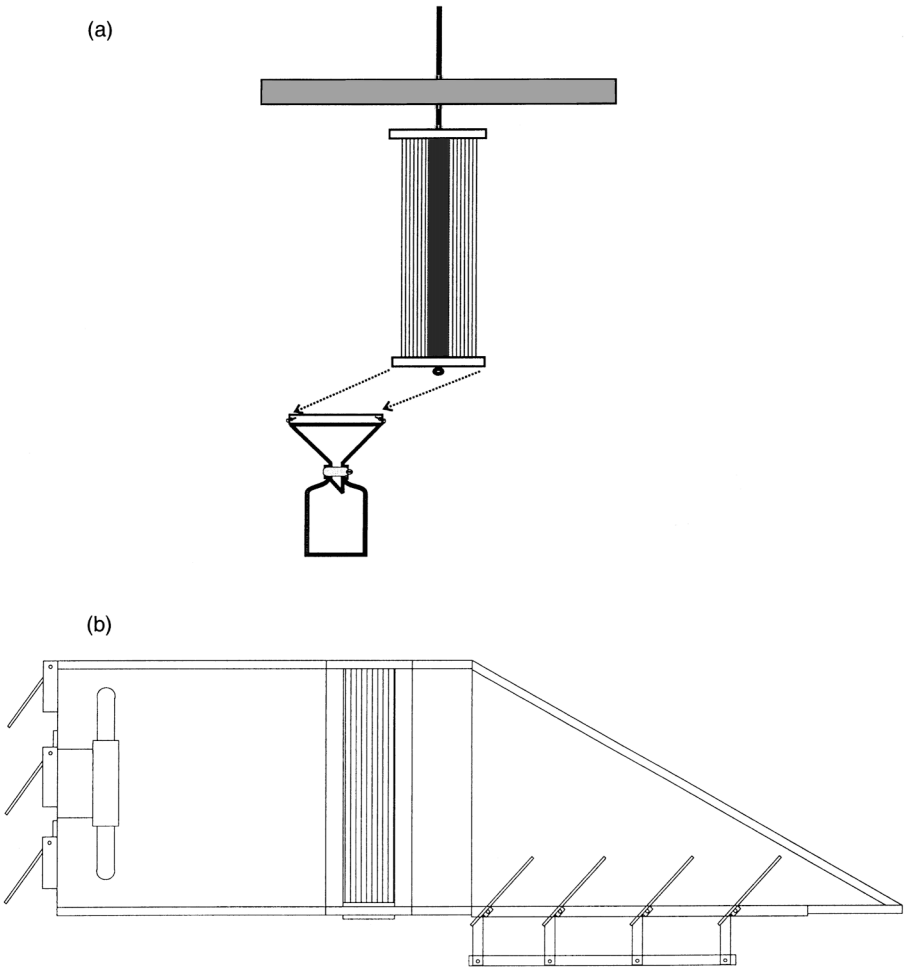
Fog collection

Fog samples were collected for approximately 1 year from passive and active collectors at CP and BF. The passive collectors were used to estimate fog water flux rates to the forest canopy. The active collectors were used to collect fog water samples for chemical analysis. The passive samplers consisted of a series of 120 ny-

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

© 2000 NRC Canada

Fig. 2. Schematic of the passive (*a*) and active (*b*) fog collectors used in this study. The active collector described by Daube et al. (1987) was modified to activate automatically during fog episodes and to exclude dry deposition (see text for details).



lon lines (31 cm long, 0.2 mm diameter) arranged in parallel. Fog collected on the lines drained into the funnel base and was collected by a polypropylene bottle connected to the funnel (Fig. 2*a*). A passive collector was placed in each of the canopies of seven mature trees at CP and at BF.

The active collectors have been used successfully in many studies and are described in Daube et al. (1987). They were designed to exclude rain and snow. A propeller-type automobile cooling fan was used to draw fog through a ventral inlet and through a bank of Teflon strands. Sufficient velocity through the strands, required for efficient collection, was achieved by reducing the internal cross-sectional area of the collector with a venturi (Daube et al. 1987). The active fog collectors (one at each site) were located ca. 3 m above ground in relatively open gaps in the forest. We modified the active collectors to exclude dry deposition and to automatically activate and collect fog samples during fog events. Dry deposition was excluded by adding louvers to the front and back openings (Fig. 2*b*). The louvers or baffles remained closed until the collector fan was activated. Air flow by the fan automatically opened the louvers and allowed fog to be drawn over the collector strings. When the fog dissipated, the fan was automatically turned off and the baffles closed until the next fog event. The addition of the louvers may have slightly increased the pressure drop through the sampler, thus raising the lower droplet size cutoff. The louvers may have also affected the passage of large fog drops into the collector. These modifications, added to prevent dry depostion, may thus result in conservative estimates of fog inputs. Photographs and descriptions of the passive and active fog collector systems have been published (Bytnerowicz et al. 1999*a*).

The actuation system for the automatic collectors was designed as follows. A relative humidity (RH) sensor relays RH data to a datalogger, which at a specified RH (80%), actuates a minifan in a horizontally positioned PVC cylinder containing a wetness sensor. When enough moisture from the air passing over the sensor accumulates on the wetness sensor, a relay signal switches on the electricity to the collector fan, which pulls air through the collector causing the baffles to open and to begin fog collection. When the wetness sensor dried and the electrical resistance increased above a predetermined threshold, the relay to the collector fan would open, shutting off the fan and ending the fog collection. Field observations of the collectors confirmed that the wetness sensor threshold provided excellent control of the collectors so that they only turned on during fog events.

Fog samples were collected from the passive and active samplers on a weekly basis when fog occurred. Weekly samples often included multiple events. Fog events did not occur every week, especially during the dry summer months. The collector strings on the active sampler were cleaned monthly with distilled water and the collection bottle replaced weekly. Fog samples collected by the active samplers (one per site) were analyzed for NO_3^- and SO_4^{2-} with ion chromatography (Dionex series 4000i, Sunnyvale, Calif.) and for NH_4^+ colorimetrically with a Technicon TRAACS 800 autoanalyzer (Tarrytown, N.Y.). For all analyses, established quality-control procedures were followed, including the use of

© 2000 NRC Canada

known standards and randomly selected duplicate analyses. Average percent analytical error for NO_3^- and NH_4^+ were generally below 1% and 5%, respectively. Fog collector blank rinse samples were taken at the beginning of the experiment and levels were minimal, near background levels.

Fog samples in the passive collectors were measured for volume to determine fog deposition rates. The amount of fog deposited to the forest during the study was calculated as a factor of ionic concentrations in fog and the fog water deposition fluxes to the passive collectors. These calculations assumed that fog deposition fluxes to the canopy were similar to fluxes to the collector strings. Based on previous studies comparing string collectors with forest canopies this seems to be a reasonable assumption (Joslin et al. 1990). The canopy surface area used to calculate ionic deposition in fog was derived from LAI estimates determined indirectly with the PAR measurements taken with the radiometer as described above. Projected (one-sided) LAI values were converted to all-sided LAI (Fassnacht et al. 1994). This yielded an LAI of 2.9 for CP and 2.5 for BF. These values are probably conservative as the Ceptometer method reportedly underestimates LAI values for mixed pine–oak forests compared to direct harvest methods (Fassnacht et al. 1994).

Results

Throughfall inputs

Precipitation during the year of throughfall collection was 1311 mm at the Crestline Fire Station weather station located about 3 km from CP. Long-term average precipitation data indicated that precipitation during the throughfall study was 33% above the long-term average. Precipitation at the BF monitoring station (Fenn and Bytnerowicz 1997) was 579 mm during the throughfall study, which is 5% below average for BF. Mean annual throughfall water inputs were 1507 mm at CP and 433 mm at BF (Fig. 3). Total throughfall deposition of NO_3^- and NH_4^+ were 5 and 14 times greater, respectively, at CP, and SO_4^{2-} deposition was 7 times greater at CP compared with BF. Total N (NO_3-N + NH_4-N) and SO_4-S deposition in throughfall averaged 18.8 and 2.9 kg·ha^{-1}·year^{-1} for the four transects at CP and 2.9 and 0.4 at BF (Fig. 3). Mean volume-weighted ionic concentrations of NO_3^-, NH_4^+, and SO_4^{2-} were 47.2, 33.5, and 11.1 μequiv.·L^{-1}, respectively, at CP compared with 37.2, 9.4, and 5.4 μequiv.·L^{-1}, respectively, at BF (Fig. 4). Ionic concentrations of NO_3^-, NH_4^+, and SO_4^{2-} at CP were 1.3, 3.5, and 2.0 times greater at CP than at BF.

At CP, ionic deposition and concentrations in throughfall varied widely between transects (Figs. 3 and 4). Throughfall water inputs also varied at CP but to a much lesser degree than ionic inputs. Throughfall precipitation amount in transect 1 was 12–37% greater than in transects 2–4 at CP. Total throughfall N deposition (NO_3^- + NH_4^+) ranged from 12.1 to 31.7 kg·ha^{-1}·year^{-1} in the four transects and S deposition ranged from 2.0 to 4.6 kg·ha^{-1}·year^{-1} (Fig. 3). Deposition was similar in transects 2–4, but throughfall deposition of the three ions in transect 1 was 2.0–2.7 times greater than the average deposition in transects 2–4. A similar pattern was evident from the ion concentration data for CP (Fig. 4).

At BF, total throughfall N deposition ranged from 0.9 to 4.2 kg·ha^{-1}·year^{-1} in the four transects and S deposition ranged from 0.2 to 0.6 kg·ha^{-1}·year^{-1}. Deposition was similar in transects 2–4, with throughfall deposition of the three ions in transects 2–4 ranging from 2.1 to 3.8 times greater than in transect 1 (Fig. 3). A similar pattern was evident from the ion concentration data (Fig. 4). Throughfall precipitation amounts varied little among the transects at BF.

Net throughfall, the combined inputs from fog and dry deposition, constituted about 85% of the total throughfall deposition of NO_3^- and NH_4^+ at CP. In the case of SO_4^{2-}, three fourths of the deposition was as net throughfall and one fourth as wet deposition in the form of rain and snow (Fig. 5). At BF the proportion of NO_3^- in net throughfall versus wet deposition was similar to CP, with one third as wet deposition and two thirds as net throughfall. However, wet deposition of NH_4^+ and SO_4^{2-} at BF was as great or slightly greater than total throughfall deposition of these same ions (Fig. 5).

Fog inputs

Ionic concentrations of fogwater (Fig. 6), fog-water deposition fluxes, fog occurrence (Fig. 7), and fog density (based on visual observations) were all higher at CP compared with BF. During a 10-month period in 1997, fog was collected from the passive collectors on 19 dates at CP but on only 9 dates at BF. The average volume of fog collected by the passive collectors was 50 mL at CP compared with 28 mL at BF (Fig. 7). Average fog deposition fluxes to the passive fog collectors during the study were 54 mL·m^{-2}·h^{-1} at CP (from February to December 1997) and 32 mL·m^{-2}·h^{-1} at BF (from November 1996 to January 1998). At CP, peak concentrations of NH_4^+, NO_3^-, and SO_4^{2-} were 1423, 1234, and 326 μequiv.·L^{-1}, resepectively. At BF, corresponding values for peak concentrations of NH_4^+, NO_3^-, and SO_4^{2-} were 254, 324, and 156 μequiv.·L^{-1}, respectively (Fig. 6). Fog samples were slightly more (p = 0.08) acidic at CP (4.2) than at BF (4.8; Fig. 8).

Annual estimated stand-level deposition of NH_4-N, NO_3-N, and SO_4-S in fog were 6.0, 4.9, and 1.9 kg·ha^{-1}, respectively, at CP. Analogous values for BF were 0.2, 0.4, and 0.2 kg·ha^{-1}, respectively. We estimated that N deposition in fog contributed 13% of the total annual N deposition at BF (0.6 kg·ha^{-1}) and 35% at CP (10.9 kg N·ha^{-1}). Analogous values for S were 26% at BF and 39% at CP. However, these estimates are based on throughfall data predominantly from 1996 and fog data from 1997. Nonetheless, these estimates provide an indication of the relative importance of fog deposition in the SBM.

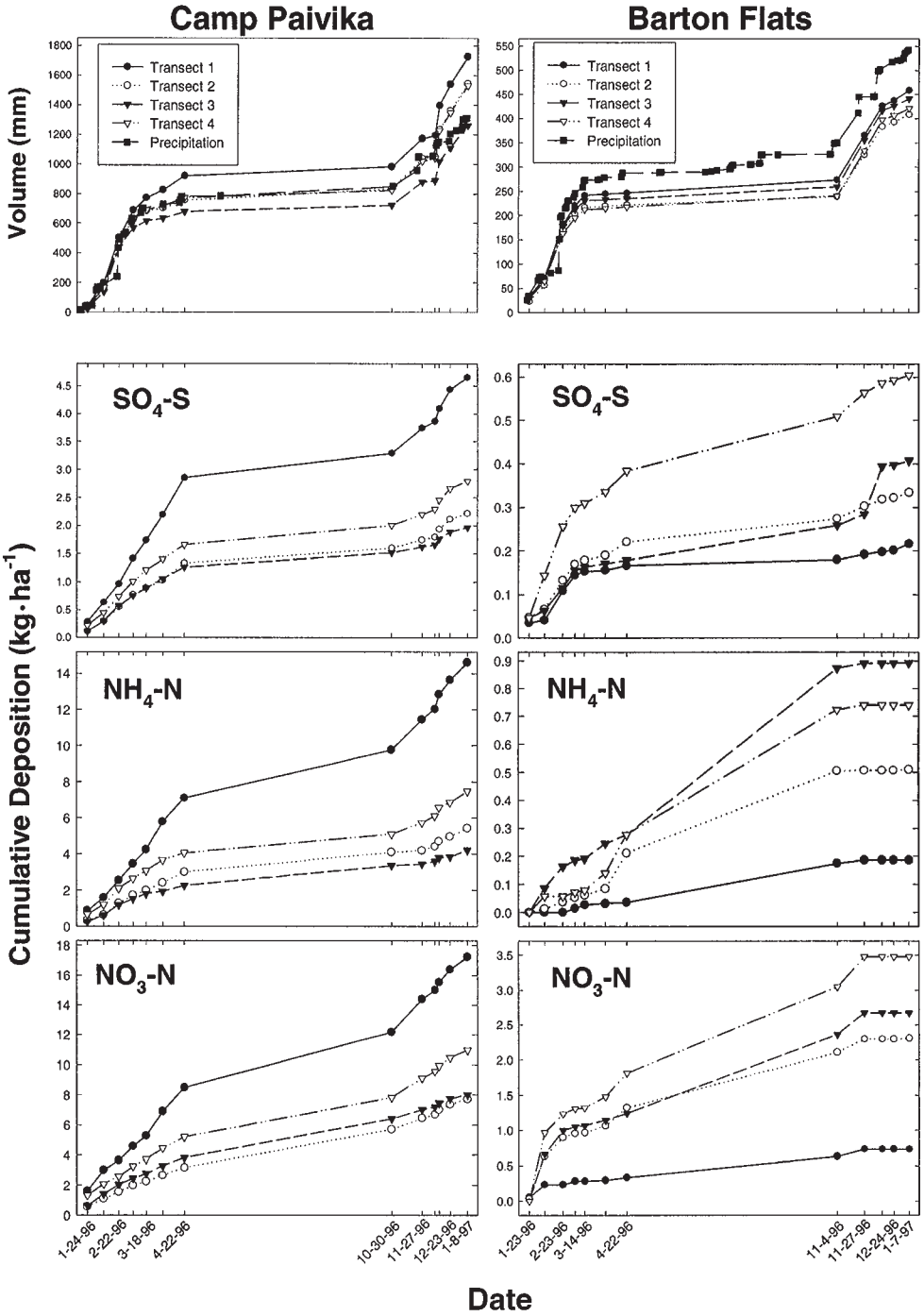
Discussion

Nitrogen deposition in the San Bernardino Mountains

Nitrogen inputs in throughfall and fog were 6.5 and 18.2 times higher at CP in the western SBM than at BF in the eastern SBM. These findings confirm earlier reports of a steep N deposition gradient in the SBM based on previous dry deposition and fog deposition measurements. Nitrogen deposition and atmospheric concentrations decline exponentially, while ozone concentrations decrease linearly along the west–east air pollution gradient in the SBM (Fenn and Bytnerowicz 1993; Miller et al. 1986). Nitrogen concentrations in the atmosphere and N deposition to the forest decrease with greater distance from the urban pollution source area to the west. The decrease in N deposition between CP and BF is dramatic considering that these sites are only

© 2000 NRC Canada

Fig. 3. Cumulative deposition of NO_3^-, NH_4^+, and SO_4^{2-} and volume of precipitation and throughfall in the four transects (eight collectors per transect) at Camp Paivika and Barton Flats.



42 km apart. Higher throughfall inputs at CP are mainly attributed to much greater air pollution exposure (Fenn and Bytnerowicz 1993). Secondary causes of greater N deposition at CP include much greater fog incidence, greater precipitation volumes (Prado-Fiedler 1990), and greater canopy leaf area.

It isn't known how the results of this study compare with the long-term average deposition inputs for the SBM, but we expect that precipitation amount and temporal dynamics of precipitation in relation to dry deposition accumulation on the forest canopy are the major factors affecting annual throughfall variation. Precipitation data from near Crestline (3 km east of CP) indicate that precipitation volumes were approximately 30% above the long-term average during the period of throughfall collection. At BF, precipitation during the throughfall study was near the long-term average. However, no precipitation or only trace amounts occurred during a 6-month period from April 22 through October 1996. This certainly reduced the amount of atmospheric deposition to the forest canopy that was collected in throughfall, compared

© 2000 NRC Canada

Fig. 4. Volume-weighted average ionic concentrations in throughfall at Camp Paivika and Barton Flats.

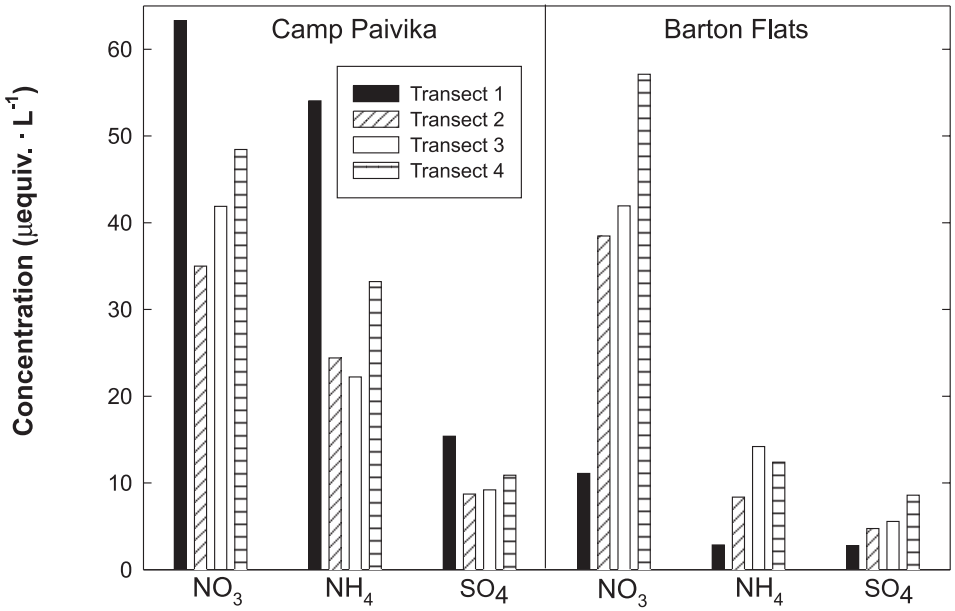
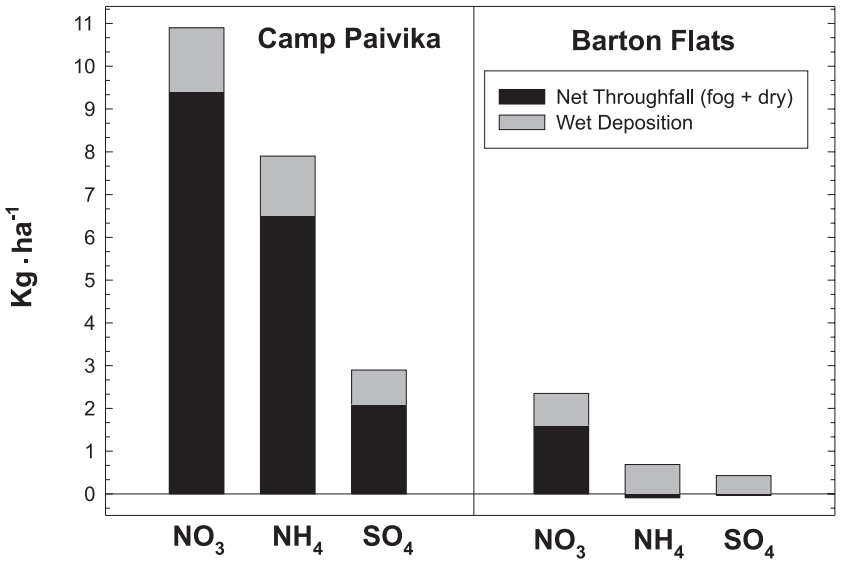


Fig. 5. Net throughfall (fog + dry deposition) and wet deposition (rain and snow) at Barton Flats and Camp Paivika (January 1996 – January 1997).



with years when infrequent summer thundershowers occur. Summer throughfall events, although typically of low volume, result in significant levels of N inputs to the forest floor, even at BF where N deposition is relatively low (Fenn and Bytnerowicz 1997).

The NO_x emissions from the SCAB decreased by 13% from 1985 to 1995, and nitrogen dioxide concentrations have decreased by 39% since 1980 (Alexis et al. 1999). These data suggest that deposition of oxidized forms of N was much greater in previous decades. Future trends in deposition of oxidized N forms may continue on a slowly decreasing trend as pollution controls improve and as older motor vehicles are replaced with newer lower emitting models. However, in the future, projected large population increases may partially offset pollution controls. Deposition of reduced N forms in the western SBM is expected to decrease in the next 10–15 years as local dairy farms, the major source of reduced N forms in the atmosphere (Luebs et al. 1973; Russell and Cass 1986), migrate north to central California.

Net throughfall (dry plus fog) and wet deposition components

Deposition of N and S at CP was clearly dominated by net throughfall composed of dry deposition and deposition in fog. Deposition in rain and snow at CP constituted only around 15% for N and 28% for S. Because the periods when fog and throughfall were collected only overlap for a short time, we were only able to estimate the proportion of net throughfall originating from fog versus dry deposition.

© 2000 NRC Canada

Fig. 6. Ionic concentrations in fog water at Camp Paivika and Barton Flats.

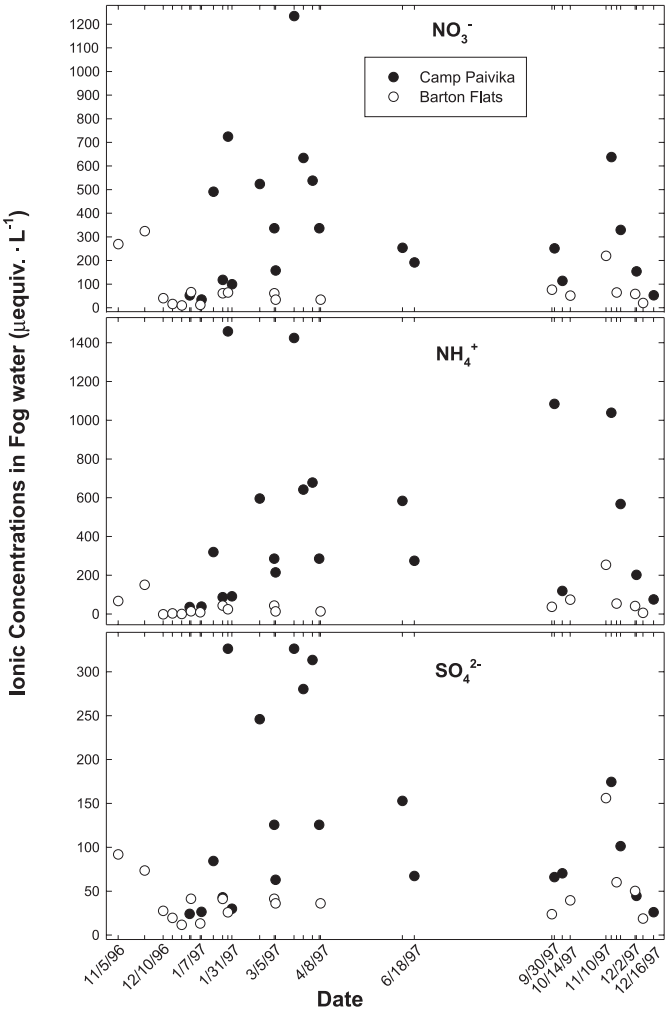


Fig. 7. Fog-water volumes collected by the passive fog collectors at Camp Paivika and Barton Flats.

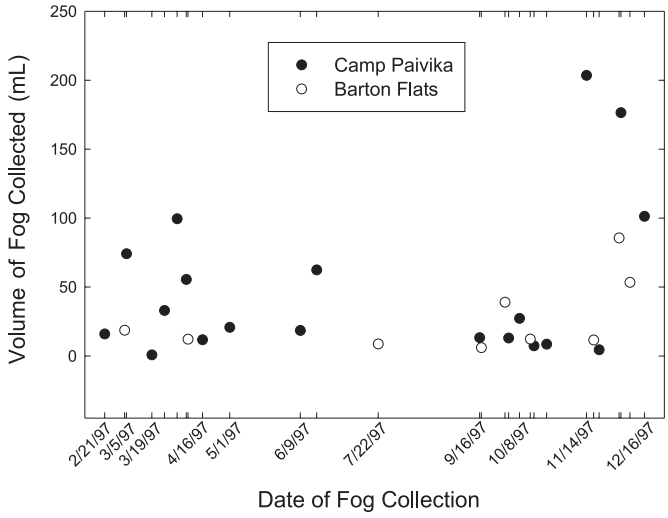
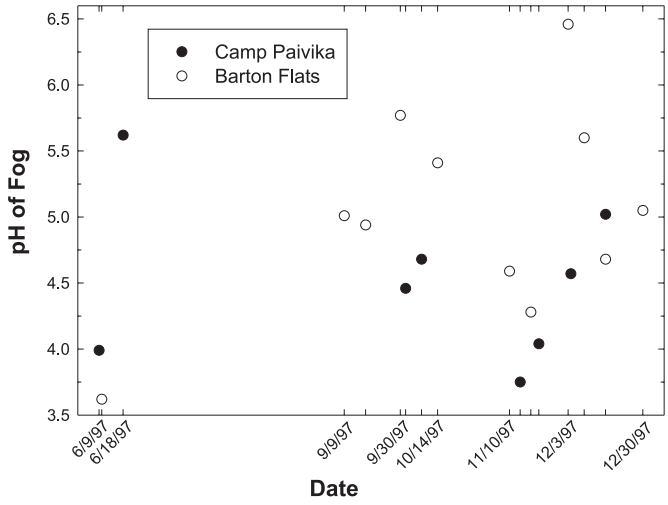


Fig. 8. Fog water pH for samples collected with the active collector.



However, our estimate of throughfall deposition originating from fog is conservative if we assume that fog inputs were even greater during the throughfall collection period when precipitation was 50% higher at CP and 40% higher at BF compared with the fog collection period. We calculated that N deposition in fog at CP was 10.9 kg·ha^{-1}·year^{-1} compared with 0.6 kg·ha^{-1}·year^{-1} at BF, values which equal 35 and 13% of estimated total N deposition (as opposed to throughfall deposition which underestimates total deposition; see discussion below) at CP and BF. These data emphasize the importance of fog in the N deposition budget at CP. Dense fog is a common occurrence at CP during portions of the fall, winter, and spring. Canopies at CP often saturate with fog until moisture drips from the trees onto the forest floor. Fog is a less common occurrence at BF, and fog-water deposition fluxes and ionic concentrations are also lower at BF compared with CP. Fog deposition of N in particular has high potential for canopy N retention because of the extremely elevated N concentrations in fog and the long residence time of the nutrient solutions saturating the foliage and branches. Fog deposition of N was estimated to be greater than N deposition in precipitation at a forested site in Sequoia National Park in the southern Sierra Nevada Mountains in central California (Collett et al. 1989), and fog or cloud-water deposition to forests has been shown to be important in a number of studies (Lovett 1994; Miller et al. 1993; Weathers et al. 1995).

At BF, NO_3^- deposition was also dominated by net throughfall inputs. However, net throughfall deposition values for NH_4^+ and SO_4^{2-} at BF were slightly negative. The most likely factors accounting for the lack of measured dry deposition of NH_4^+ and SO_4^{2-} at BF include low deposition rates of NH_4^+ and SO_4^{2-} in fog and in dry forms (Fenn and Bytnerowicz 1993; Fenn and Bytnerowicz 1997), and the 6-month dry period when only slight fog deposition occurred and no throughfall was collected. Although speculative, it may also be that canopy retention of atmospheric ions is more effective at BF than at CP because of a greater number of available adsorption sites, thus resulting in greater canopy ion retention and lower net throughfall estimates. In contrast, the high deposition rates at CP may saturate canopy adsorption sites more rapidly, contributing to higher net throughfall values.

© 2000 NRC Canada

Table 1. Estimated projected leaf area index (one sided) and inorganic N and S deposition ($kg \cdot ha^{-1} \cdot year^{-1}$) in throughfall in the four transects at Camp Paivika and Barton Flats.

Transect	Projected LAI	N deposition*	SO_4-S deposition
Camp Paivika			
1	1.38	31.8	4.6
2	1.20	13.1	2.2
3	0.94	12.2	1.9
4	3.03	18.3	2.8
Barton Flats			
1	0.40	0.9	0.2
2	0.98	2.8	0.3
3	2.13	3.6	0.4
4	1.54	4.2	0.6

Note: Projected LAI was estimated indirectly with an integrating radiometer. See text for details.

*Sum of NO_3^- and NH_4^+ deposition.

Spatial variability in throughfall deposition

Within-site spatial variability in throughfall deposition fluxes and ionic concentrations is evident from comparisons between the four transects at each site. At BF, transect 1 was in an area of the stand with large open gaps in the canopy. Indirect estimates of LAI, derived from measurements of photosynthetically active radiation (Pierce and Running 1988), were 2.5–5.3 times lower in transect 1 at BF compared with the other three transects (Table 1). As a result, ionic concentrations and total deposition in transect 1 at BF were much lower than in the other transects. At CP, ionic concentrations and deposition were much greater in transect 1 than in transects 2–4. However, differences in canopy coverage among transects do not explain the higher deposition in transect 1 at CP (Table 1), and throughfall volume was only slightly greater in transect 1 compared with the other three transects. Greater exposure to the incoming air pollution from the west seems to be the most likely cause of the greater deposition in transect 1 at CP. Transect 1 has the most direct exposure to Devil Canyon where pollutants are transported to the stand from the urban smog sources in the valley directly below. An effect of leaf area on throughfall deposition was apparent among transects 2–4 at CP, however (Table 1). Transect 4 has the least direct exposure to incoming polluted air masses at CP, yet among transects 2–4, deposition of N and S and LAI were highest in transect 4. Nitrogen deposition ($NH_4^+ + NO_3^-$) was significantly correlated with LAI at both study sites ($r = 0.99$ at CP excluding transect 1; $r = 0.85$ at BF).

Throughfall inorganic N deposition for the four transects at CP ranged from 12 to 32 $kg \cdot ha^{-1} \cdot year^{-1}$. The highest throughfall deposition is expected in microsites where complete canopy cover, strong edge effects (Weathers et al. 1995; Draaijers et al. 1988; Lindberg and Owens 1993), and other exposure characteristics favor elevated atmospheric deposition. The high degree of variability in atmospheric deposition across the landscape is analogous to applying varying levels of fertilization to sites within the stand. Availability of nutrients in throughfall to individual trees or groups of trees in the vicinity will be a function of subsequent processing and hydrologic transport of nutrients within the forest floor and underlying rooting zone.

Variable deposition rates throughout the stand are to be expected considering the open nature of Western mixed conifer forests compared with forests with closed canopies. At the BF throughfall study site, stand cover is 67% (Fenn and Bytnerowicz 1997). Percent stand cover is slightly higher at CP based on visual observations and below-canopy PAR measurements. Stand-level deposition fluxes given in this paper include deposition to open areas, meaning that deposition rates under tree canopies are greater than average stand-level rates. The widespread occurrence of gaps in these stands leads to high spatial variability in N inputs, because open areas have little capacity to intercept atmospheric pollutants. The large number of gaps also creates extensive areas of the forest where edge effects affect deposition (Weathers et al. 1995; Draaijers et al. 1988; Lindberg and Owens 1993). Average deposition inputs are useful for stand-level N budgets, but values may grossly misrepresent the range of deposition inputs that occur at smaller scales (e.g., under individual trees or small clusters of trees). Nitrogen deposition rates at CP in "hotspots of deposition" (Weathers et al. 2000), such as under tree canopies in more directly exposed areas, are possibly within the range of 40–50 $kg \cdot ha^{-1} \cdot year^{-1}$ or more.

It should also be noted that stemflow was not measured and that canopy throughfall inputs in this and most studies do not include deposition to the forest floor or to understory vegetation. At CP the understory is dominated by dense bracken fern growth. Because of the high leaf area of the understory at CP, deposition fluxes to the understory are expected to be a significant component of the overall N deposition budget; however, they were not measured in this study. The understory at BF is relatively sparse except for some areas where thickets of white fir saplings occur.

In spite of the large differences in throughfall deposition of N, and to a lesser degree of S, between CP and BF, NO_3^- concentrations in throughfall were not statistically different at CP and BF. Similarly, NH_4^+ and SO_4^{2-} concentrations at the two sites differed little compared with total deposition amounts. As previously reported (Fenn and Bytnerowicz 1997), ionic concentrations in throughfall at BF were high relative to deposition rates, because of the low throughfall volumes. Throughfall volumes at BF (433 mm annual throughfall) were on average only 29% as great as at CP (1507 mm).

Sources and forms of N deposition

In southern California, where fossil fuel consumption is by far the primary source of nitrogenous air pollution, oxidized forms of N predominate (Bytnerowicz and Fenn 1996; Bytnerowicz et al. 1987*b*). However, in areas with significant agricultural activity, emissions and deposition of reduced forms of N increase. This has been documented at Whitaker Forest, adjacent to Sequoia National Park, in the southwestern Sierra Nevada, presumably as a result of the large agricultural region in the Central Valley of California (Bytnerowicz and Riechers 1995) and also at Camp Paivika in this and a previous study (Fenn and Bytnerowicz 1997). Deposition of NH_4^+ in fog and throughfall were 122 and 72% as high as NO_3^- deposition at Camp Paivika. The high

© 2000 NRC Canada

NH_4^+ inputs at CP are due to emissions of NH_3 from the highly concentrated dairy industry in the Chino–Norco area 34 km southwest of CP (Luebs et al. 1973) Although not yet studied in the SBM, throughfall deposition of organic N compounds may be another important deposition form and source of N inputs to the soil. Recent studies suggest that deposition inputs and leaching losses of organic N are significant components of the N budget in many ecosystems (Crockford et al. 1996; Currie et al. 1996).

Throughfall deposition versus total deposition

In areas of chronic N deposition, throughfall underestimates total N deposition because of canopy retention of atmospheric N (Lovett and Lindberg 1993). The dominant mechanisms of canopy N retention are not well defined, but include physical adsorption, stomatal or transcuticular uptake by plant tissue, and uptake by epiphytes and plant-associated microbes (Huttunen 1996). Nitric acid vapor (HNO_3) is one of the predominant nitrogenous pollutants at CP (Fenn and Bytnerowicz 1993), and the high deposition velocity of HNO_3 underscores its importance in the N deposition budget (Hanson and Lindberg 1991). Nitric acid vapor exposure has been shown to induce nitrate reductase activity in foliage of ponderosa pine and two associated oak species (Krywult and Bytnerowicz 1997), thus providing evidence of foliar uptake. Transcuticular transport of $^{15}HNO_3$ into the leaf interior has also been demonstrated for ponderosa pine and California black oak (Bytnerowicz et al. 1998). Stomatal uptake of nitrogenous pollutants also is expected (Stulen et al. 1998; Wellburn 1998). In summary, while our understanding of the relative importance of canopy retention mechanisms for nitrogenous pollutants is incomplete, it is clear that canopy retention of atmospheric N causes throughfall to underestimate total N deposition by a significant degree.

Empirical results from the Integrated Forest Study (IFS) sites (Johnson and Lindberg 1992), where total deposition and throughfall deposition of N and S were extensively monitored, demonstrated that only about 60% of the total N deposition was measured as throughfall N deposited to the forest floor (Lovett 1994; Lovett and Lindberg 1993). Canopy retention of dry-deposited N is much greater than for wet-deposited N (Boyce et al. 1996; Fenn and Leininger 1995; Garten et al. 1998; Lovett and Lindberg 1993). At Whiteface Mountain in New York, 32% of the N deposited to the forest canopy occurred as cloud water and 25% as HNO_3; these deposition proportions are similar to N deposition at CP. At Whiteface Mountain, only 47% of N deposition was measured in throughfall (Miller et al. 1993). Canopy retention of atmospheric N is likely to be similarly significant at CP, especially considering that a large fraction of the inorganic N deposition at CP occurs as NH_4^+ which is generally more effectively retained by forest canopies than NO_3^- (Lovett 1994). On the other hand, canopy retention of S is considered to be minimal, and throughfall flux of S is reportedly a good surrogate for total S deposition, especially when stemflow is also measured (Joslin and Wolfe 1992; Butler and Likens 1995; Lovett 1994; Lovett and Lindberg 1993; Miller et al. 1993).

The data in this study suggest that extended dry periods without throughfall, a regular occurrence in forests with a Mediterranean climate, further complicate the interpretation of throughfall data. Questions arise regarding the relationship between throughfall deposition and total deposition fluxes during these extended rain-free periods. The cumulative throughfall deposition data for April 1996 through November 1996 (when air pollution levels were highest) shown in Fig. 3 strongly suggest that a large portion of the dry-deposited N and S, which normally would have been collected as throughfall if it had rained, is not accounted for in subsequent precipitation events. Using previously published values for dry deposition fluxes of NO_3^-, NH_4^+, and SO_4^{2-} to pine branches at our study sites based on successive bi-weekly branch rinses (Fenn and Bytnerowicz 1993), we calculated the amount of inorganic N and S deposited to the forest canopy in dry forms during the 6-month dry period and to the end of the two subsequent throughfall collection dates (Nov. 27, 1996; a total of 7 months). The throughfall collected in November underestimated the N deposition that would have been collected by periodic throughfall events during the previous 7 months by ca. 50% at both sites compared with deposition estimates from branch rinse data. We also calculated that S deposition was underestimated by ca. 50% at CP and by 75% at BF. Thus, during and after these smoggy drought periods, throughfall severely underestimates total atmospheric deposition. We assume, however, that during the winter wet season, canopy retention of deposited ions and throughfall underestimation of deposition in the SBM was similar to that in more mesic forests of similar vegetation types (Lovett and Lindberg 1993).

The degree of underestimation in throughfall following long dry periods is quite surprising, especially for S, and we can only speculate as to the fate of the unaccounted for N and S. The most likely sinks for the N and S not collected as throughfall would include plant uptake and adsorption, volatilization losses, dry fallout of particles, and microbial uptake, although the latter is expected to be be minimal during the long dry summers. The mechanisms of deposition, pollutant interactions with canopy surfaces, and interpretation of throughfall data in Mediterranean climates is clearly an area for future research.

If we conservatively assume that canopy interactions with atmospheric N behave similarly in the SBM as in the IFS sites (i.e., that annual throughfall deposition underestimates inorganic N deposition by approximately 40%), this results in estimated inorganic N deposition of 20–53 $kg \cdot ha^{-1} \cdot year^{-1}$ in the four transects at CP, and 1.5–7 $kg \cdot ha^{-1} \cdot year^{-1}$ at BF. Accordingly, plot-level average values for inorganic N deposition would be 31 $kg \cdot ha^{-1} \cdot year^{-1}$ at CP and 5 $kg \cdot ha^{-1} \cdot year^{-1}$ at BF. If we assume that S deposition was also underestimated by 40%, then total estimated S deposition would be 3.3–7.7 $kg \cdot ha^{-1} \cdot year^{-1}$ in the four transects at CP and 0.4–1.0 $kg \cdot ha^{-1} \cdot year^{-1}$ at BF. Plot-level averages for estimated total S deposition would be 4.8 $kg \cdot ha^{-1} \cdot year^{-1}$ at CP and 0.6 $kg \cdot ha^{-1} \cdot year^{-1}$ at BF. Of course, these estimates entail a high degree of uncertainty and serve only for discussion and to illustrate approximate ranges for N and S loadings at CP and BF if throughfall underestimates total atmospheric inputs as previous studies and dry deposition flux calculations suggest.

Total deposition inputs in this study were also underestimated, because stemflow was not measured. However,

© 2000 NRC Canada

stemflow is not expected to be a large component of the total deposition in these stands, which have a low stand cover of approximately 60–70% and with only a small fraction of very young trees. Stemflow is an important ionic input for smooth-barked species (including young trees) and stands with a high stem density (Parker 1983; Thimonier 1998). In the IFS it was found that, in sites with very high stem density, stemflow deposition was 6–10% of total deposition; otherwise, stemflow deposition was less than 2% of the total (Johnson 1992). Stemflow is commonly less than 3% of the total input in mature pine and oak forests but has been reported to be 10% or more in young stands (Thimonier 1998). In summary, the characteristics of the stands at CP and BF suggest that stemflow is likely to be much less than 5% of the total deposition.

Throughfall deposition thresholds for N saturation

An important reason for quantifying N deposition in forests is to increase our ability to predict the levels of N deposition at which ecological and environmental impacts become significant. In a broad survey of 65 watersheds in Europe it was reported that below a throughfall deposition threshold of 10 kg N $ha^{-1}\cdot year^{-1}$, no significant NO_3^- leaching from the forest occurred, while at intermediate levels of 10–25 kg N $ha^{-1}\cdot year^{-1}$, leaching occurred at some sites. Above a throughfall deposition level of 25 kg N $ha^{-1}\cdot year^{-1}$, N leaching occurred at all sites (Dise and Wright 1995). Ecosystems in the San Bernardino and San Gabriel Mountains in southern California support this general finding, notwithstanding the widely different climatic, biological, and physical characteristics of watersheds in California compared with central Europe. A high-elevation mixed conifer forest in the San Gabriel Mountains where throughfall deposition of N was 11.5 $kg\cdot ha^{-1}\cdot year^{-1}$ was N limited based on responses of Jeffrey pine trees to N fertilization (Kiefer and Fenn 1997). On the other hand, the forest at CP is clearly N saturated as evidenced by a number of botanic and edaphic N-status indicators (Fenn et al. 1996) and from spatial and temporal trends in stream water NO_3^- concentrations (Fenn and Poth 1999). Average NO_3^- concentrations were 151 and 65 $\mu equiv.\cdot L^{-1}$ near the top and bottom ends, respectively, of one of the main streams in Devil Canyon (the West Fork), located immediately west and downslope of CP. Stream water NO_3^- concentrations were also highly elevated in chaparral watersheds in the San Gabriel Mountains, where throughfall N deposition was reported to be 23 $kg\cdot ha^{-1}\cdot year^{-1}$ (Riggan et al. 1985, 1994).

Summary

Although only 42 km apart, N deposition in throughfall decreased sharply from CP to BF (from 18.8 to 2.9 $kg\cdot ha^{-1}\cdot year^{-1}$), and S deposition decreased from 2.9 $kg\cdot ha^{-1}\cdot year^{-1}$ at CP to 0.4 $kg\cdot ha^{-1}\cdot year^{-1}$ at BF. Estimated annual fog deposition of N and S at CP were 10.9 and 1.9 $kg\cdot ha^{-1}$, respectively, compared with 0.6 and 0.2 $kg\cdot ha^{-1}$ at BF. Ionic concentrations of fog water, fog-water deposition fluxes, fog occurrence, and fog density were all higher at CP compared with BF. Ionic concentrations and annual deposition of N and S in throughfall were variable among the four transects within each site, presumably because of varying exposure to pollutant-bearing air masses and differences in the amount of leaf area above the throughfall collectors. The results of this study illustrate the highly variable spatial distribution of atmospheric deposition within these heterogenous and relatively open stands. In this summer-dry climate, dry-deposited pollutants accumulate on canopy surfaces and on the forest floor during long precipitation-free periods, followed by concentrated pulses of throughfall washoff during precipitation events. Ionic concentrations and annual deposition of N can be particularly elevated in more directly exposed areas under mature trees with large canopy surface area and in microsites of complete canopy cover (Fenn and Bytnerowicz 1997).

Based on previous studies, total deposition of N to the forest canopy may have been underestimated by 40% or more in throughfall measurements due to canopy retention of atmospherically deposited N. In addition, a significant amount of N and S deposited to the forest canopy during a 6-month dry period concurrent with the peak smog season was not accounted for in subsequent throughfall collections. The results of this and other recent studies of N deposition in the transverse ranges of southern California (Fenn and Kiefer 1999) suggest that throughfall N deposition thresholds at the landscape scale (e.g., per hectare) at which elevated NO_3^- losses and other symptoms of N saturation occur are similar to thresholds reported for forested watersheds in Europe (Dise and Wright 1995). Nitrogen deposition levels in the western SBM are also sufficient to exert dramatic physiological effects on trees and to significantly alter carbon and nitrogen cycling (Takemoto et al. 2000). Further mechanistic studies are needed to elucidate the interactions of atmospheric pollutants with forest canopies and to improve monitoring methods for estimation of total atmospheric deposition inputs in these summer-dry wildland ecosystems.

Acknowledgments

We thank Dave Jones and Tony Gomez for invaluable assistance in installing the fog and throughfall collectors at the field sites and for collecting samples during the course of the study. The excellent chemical analysis work of Diane Alexander, Karlos Rueckert, Enrique Jimenez, and Dave Hamilton are also gratefully acknowledged. We also thank Dr. Brent Takemoto for his guidance and support and the California Air Resources Board for funding this research. The generosity of Kermit Cromack and Kathy Weathers in loaning the active fog collectors for this study and the helpful reviews of an earlier draft of this manuscript by Pam Padgett and Kathy Weathers are also much appreciated.

References

Alexis, A., Gaffney, P., Garcia, C., Nystrom, M., and Rood, R. 1999. The 1999 California Almanac of Emissions and Air Quality. California Environmental Protection Agency, Air Resources Board. Sacramento, Calif.

Arbaugh, M.J., Johnson, D.W., and Pulliam, W.M. 1999. Simulated effects of N deposition, ozone injury and climate change on a forest stand in the San Bernardino Mountains. *In* Oxidant air pollution impacts in the montane forests of southern California:

© 2000 NRC Canada

a case study of the San Bernardino Mountains. *Edited by* P.R. Miller and J.R. McBride. Ecol. Stud. 134. pp. 353–372.

Boyce, R.L., Friedland, A.J., Chamberlain, C.P., and Poulson, S.R. 1996. Direct canopy nitrogen uptake from ^{15}N-labeled wet deposition by mature red spruce. Can. J. For. Res. **26**: 1539–1547.

Butler, T.J., and Likens, G.E. 1995. A direct comparison of throughfall plus stemflow to estimates of dry and total deposition for sulfur and nitrogen. Atmos. Environ. **29**: 1253–1265.

Bytnerowicz, A., and Fenn, M.E. 1996. Nitrogen deposition in California forests: a review. Environ. Pollut. **92**: 127–146.

Bytnerowicz, A., and Riechers, G. 1995. Nitrogenous air pollutants in a mixed conifer stand of the western Sierra Nevada, California. Atmos. Environ. **29**: 1369–1377.

Bytnerowicz, A., Miller, P.R., and Olszyk, D.M. 1987*a*. Dry deposition of nitrate, ammonium and sulfate to a *Ceanothus crassifolius* canopy and surrogate surfaces. Atmos. Environ. **21**: 1749–1757.

Bytnerowicz, A., Miller, P.R., Olszyk, D.M., Dawson, P.J., and Fox, C.A. 1987*b*. Gaseous and particulate air pollution in the San Gabriel Mountains of southern California. Atmos. Environ. **21**: 1805–1814.

Bytnerowicz, A., Percy, K., Riechers, G., Padgett, P., and Krywult, M. 1998. Nitric acid vapor effects on forest trees—deposition and cuticular changes. Chemosphere, **36**: 697–702.

Bytnerowicz, A., Carroll, J.J., Takemoto, B.K., Miller, P.R., Fenn, M.E., and Musselman, R.C. 1999*a*. Distribution and transport of air pollutants to vulnerable California ecosystems. *In* Integrated assessment of ecosystem health. *Edited by* K.M. Scow, G.E. Fogg, D.E. Hinton, and M.L. Johnson. Lewis Publishers, Boca Raton, Fla. pp. 93–118.

Bytnerowicz, A., Fenn, M.E., Miller, P.R., and Arbaugh, M.J. 1999*b*. Wet and dry pollutant deposition to the mixed conifer forest. *In* Oxidant air pollution impacts in the montane forests of southern California: a case study of the San Bernardino Mountains. *Edited by* P.R. Miller and J.R. McBride. Ecol. Stud. 134. pp. 235–269.

Collett, J., Jr., Daube, B., Jr., Munger, W.J., and Hoffmann, M.R. 1989. Cloudwater chemistry in Sequoia National Park. Atmos. Environ. **23**: 999–1007.

Crockford, R.H., Richardson, D.P., and Sageman, R. 1996. Chemistry of rainfall, throughfall and stemflow in a eucalypt forest and a pine plantation in south-eastern Australia: 1. Rainfall. Hydrol. Process, **10**: 1–11.

Currie, W.S., Aber, J.D., McDowell, W.H., Boone, R.D., and Magill, A.H. 1996. Vertical transport of dissolved organic C and N under long-term N amendments in pine and hardwood forests. Biogeochemistry, **35**: 471–505.

Daube, B., Jr., Kimball, K.D., Lamar, P.A., and Weathers, K.C. 1987. Two new ground-level cloud water sampler designs which reduce rain contamination. Atmos. Environ. **21**: 893–900.

Dise, N.B., and Wright, R.F. 1995. Nitrogen leaching from European forests in relation to nitrogen deposition. For. Ecol. Manage. **71**: 153–161.

Draaijers, G.P.J., Ivens, W.P.M.F., and Bleuten, W. 1988. Atmospheric deposition in forest edges measured by monitoring canopy throughfall. Water Air Soil Pollut. **42**: 129–136.

Fassnacht, K.S., Gower, S.T., Norman, J.M., and Mcmurtrie, R.E. 1994. A comparison of optical and direct methods for estimating foliage surface area index in forests. Agric. For. Meteorol. **71**: 183–207.

Fenn, M.E., and Bytnerowicz, A. 1993. Dry deposition of nitrogen and sulfur to ponderosa and Jeffrey pine in the San Bernardino National Forest in southern California. Environ. Pollut. **81**: 277–285.

Fenn, M.E., and Bytnerowicz, A. 1997. Summer throughfall and winter deposition in the San Bernardino Mountains in southern California. Atmos. Environ. **31**: 673–683.

Fenn, M.E., and Kiefer, J.W. 1999. Throughfall deposition of nitrogen and sulfur in a Jeffrey pine forest in the San Gabriel Mountains, southern California. Environ. Pollut. **104**: 179–187.

Fenn, M.E., and Leininger, T.D. 1995. Uptake and distribution of nitrogen from acidic fog within a ponderosa pine (*Pinus ponderosa* Laws.) litter/soil system. For. Sci. **41**: 645–663.

Fenn, M.E., and Poth, M.A. 1999. Temporal and spatial trends in streamwater nitrate concentrations in the San Bernardino Mountains, southern California. J. Environ. Qual. **28**: 822–836.

Fenn, M.E., Poth, M.A., and Johnson, D.W. 1996. Evidence for nitrogen saturation in the San Bernardino Mountains in southern California. For. Ecol. Manage. **82**: 211–230.

Fenn, M.E., Poth, M.A., Aber, J.D., Baron, J.S., Bormann, B.T., Johnson, D.W., Lemly, A.D., McNulty, S.G., Ryan, D.F., and Stottlemyer, R. 1998. Nitrogen excess in North American ecosystems: predisposing factors, ecosystem responses, and management strategies. Ecol. Appl. **8**: 706–733.

Garten, C.T., Schwab, A.B., and Shirshac, T.L. 1998. Foliar retention of ^{15}N tracers: implications for net canopy exchange in low- and high-elevation forest ecosystems. For. Ecol. Manage. **103**: 211–216.

Grulke, N.E., and Balduman, L. 1999. Deciduous conifers: high N deposition and O_3 exposure effects on growth and biomass allocation in ponderosa pine. Water Air Soil Pollut. **116**: 235–248.

Grulke, N.E., Andersen, C.P., Fenn, M.E., and Miller, P.R. 1998. Ozone exposure and nitrogen deposition lowers root biomass of ponderosa pine in the San Bernardino Mountains, California. Environ. Pollut. **103** : 63–73.

Hanson, P.J., and Lindberg, S.E. 1991. Dry deposition of reactive nitrogen compounds: a review of leaf, canopy and non-foliar measurements. Atmos. Environ. **25A**: 1615–1634.

Huttunen, S. 1996. Interactions between epiphytic microbes and deposited compounds. *In* Plant cuticles. *Edited by* G. Kerstiens. BIOS Scientific Publishers Ltd. Oxford, U.K. pp. 301–317.

Johnson, D.W. 1992. Base cations. *In* Atmospheric deposition and forest nutrient cycling. *Edited by* D.W. Johnson and S.E. Lindberg. Ecol. Stud. 91. pp. 233–340.

Johnson, D.W., and Lindberg, S.E. 1992. Atmospheric deposition and forest nutrient cycling. Ecol. Stud. 91.

Joslin, J.D., and Wolfe, M.H. 1992. Tests of the use of net throughfall sulfate to estimate dry and occult sulfur deposition. Atmos. Environ. **26**: 63–72.

Joslin, J.D., Mueller, S.F., and Wolfe, M.H. 1990. Tests of models of cloudwater deposition to forest canopies using artificial and living collectors. Atmos. Environ. **24A**: 3007–3019.

Kiefer, J.W., and Fenn, M.E. 1997. Using vector analysis to assess nitrogen status of ponderosa and Jeffrey pine along deposition gradients in forests of southern California. For. Ecol. Manage. **94**: 47–59.

Krywult, M., and Bytnerowicz, A. 1997. Induction of nitrate reductase activity by nitric acid vapor in California black oak (*Quercus kelloggii*), canyon live oak (*Quercus chrysolepis*), and ponderosa pine (*Pinus ponderosa*) seedlings. Can. J. For. Res. **27**: 2101–2104.

Lindberg, S.E., and Owens, J.G. 1993. Throughfall studies of deposition to forest edges and gaps in montane ecosystems. Biogeochemistry, **19**: 173–194.

Lovett, G.M. 1994. Atmospheric deposition of nutrients and pollutants in North America: an ecological perspective. Ecol. Appl. **4**: 629–650.

© 2000 NRC Canada

Lovett, G.M., and Lindberg, S.E. 1993. Atmospheric deposition and canopy interactions of nitrogen in forests. Can. J. For. Res. **23**: 1603–1616.

Lovett, G.M., Bowser, J.J., and Edgerton, E.S. 1997. Atmospheric deposition to watersheds in complex terrain. Hydrol. Process, **11**: 645–654.

Lovett, G.M., Weathers, K.C., and Sobczak, W.V. 2000. Nitrogen saturation and retention in forested watersheds of the Catskill Mountains, New York. Ecol. Appl. **10**: 73–84.

Luebs, R.E., Davis, K.R., and Laag, A.E. 1973. Enrichment of the atmosphere with nitrogen compounds volatilized from a large dairy area. J. Environ. Qual. **2**: 137–141.

Miller, P.R., Taylor, O.C., and Poe, M.P. 1986. Spatial variation of summer ozone concentrations in the San Bernardino Mountains. *In* Proceedings of the Air Pollution Control Association Annual Meeting, 22–27 June 1986, Minneapolis, Minn. Air Pollution Control Association, Pittsburgh, Pa. pp. 3:86–39.2.

Miller, E.K., Panek, J.A., Friedland, A.J., Kadlecek, J., and Mohnen, V.A. 1993. Atmospheric deposition to a high-elevation forest at Whiteface Mountain, New York, USA. Tellus, **45B**: 209–227.

Murdoch, P.S., and Stoddard, J.L. 1992. The role of nitrate in the acidification of streams in the Catskill Mountains of New York. Water Resour. Res. **28**: 2707–2720.

Parker, G.G. 1983. Throughfall and stemflow in the forest nutrient cycle. Adv. Ecol. Res. **13**: pp. 57–133.

Pierce, L.L., and Running, S.W. 1988. Rapid estimation of coniferous forest leaf area index using a portable integrating radiometer. Ecology, **69**: 1762–1767.

Prado-Fiedler, R. 1990. On the relationship between precipitation amount and wet deposition of nitrate and ammonium. Atmos. Environ. **24A**: 3061–3065.

Riggan, P.J., Lockwood, R.N., and Lopez, E.N. 1985. Deposition and processing of airborne nitrogen pollutants in Mediterranean-type ecosystems of southern California. Environ. Sci. Technol. **19**: 781–789.

Riggan, P.J., Lockwood, R.N., Jacks, P.M., Colver, C.G., Weirich, F., Debano, L.F., and Brass, J.A. 1994. Effects of fire severity on nitrate mobilization in watersheds subject to chronic atmospheric deposition. Environ. Sci. Technol. **28**: 369–375.

Russell, A.G., and Cass, G.R. 1986. Verification of a mathematical model for aerosol nitrate and nitric acid formation and its use for control measure evaluation. Atmos. Environ. **20**: 2011–2025.

Stoddard, J.L. 1994. Long-term changes in watershed retention of nitrogen: its causes and aquatic consequences. *In* Environmental chemistry of lakes and reservoirs. *Edited by* L.A. Baker. Adv. Chem. Ser. 237. pp. 223–284.

Stulen, I., Perez-Soba, M., DeKok, L.J., and Van der Eerden, L. 1998. Impact of gaseous nitrogen deposition on plant functioning. New Phytol. **139**: 61–70.

Takemoto, B.K., Bytnerowicz, A., and Fenn, M.E. 2000. Current and future effects of ozone and atmospheric nitrogen deposition on California's mixed conifer forests. For. Ecol. Manage. In press.

Thimonier, A. 1998. Measurement of atmospheric deposition under forest canopies: some recommendations for equipment and sampling design. Environ. Monitor. Assess. **52**: 353–387.

Vitousek, P.M., Aber, J.D., Howarth, R.W., Likens, G.E., Matson, P.A., Schindler, D.W., Schlesinger, W.H., and Tilman, D.G. 1997. Human alteration of the global nitrogen cycle: sources and consequences. Ecol. Appl. **7**: 737–750.

Waldman, J.M., Munger, J.W., Jacob, D.J., and Hoffmann, M.R. 1985. Chemical characterization of stratus cloudwater and its role as a vector for pollutant deposition in a Los Angeles pine forest. Tellus, **37B**: 91–108.

Weathers, K.C., Lovett, G.M., and Likens, G.E. 1995. Cloud deposition to a spruce forest edge. Atmos. Environ. **29**: 665–672.

Weathers, K.C., Lovett, G.M., Likens, G.E., and Lathrop, R. 2000. The effect of landscape features on deposition to Hunter Mountain, Catskill Mountains, New York. Ecol. Appl. **10**: 528–540.

Wellburn, A.R. 1998. Atmospheric nitrogenous compounds and ozone—is NO_x fixation by plants a possible solution? New Phytol. **139**: 5–9.

© 2000 NRC Canada