

Concentrations and Deposition of Nitrogenous Air Pollutants in a Ponderosa/Jeffrey Pine Canopy¹

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Abstract

*Nitrogenous (N) air pollutant concentrations and surface deposition of nitrate (NO_3^-) and ammonium (NH_4^+) to branches of ponderosa pine (*Pinus ponderosa* Dougl. ex. Laws.) seedlings were measured on a vertical transect in a mature ponderosa/Jeffrey (*Pinus jeffreyi* Grev. & Balf.) pine canopy in the mixed conifer forest stand in the San Bernardino Mountains of southern California during the 1992 to 1994 summer seasons (mid-May to mid-October periods). In general, concentrations of nitric acid vapor (HNO_3), nitrous acid vapor (HNO_2), ammonia (NH_3), particulate NO_3^- and NH_4^+ were elevated and not affected by the position within the canopy. In contrast, surface deposition of NO_3^- and NH_4^+ to ponderosa pine seedling branches strongly depended on position in the canopy. The highest deposition of the two ions occurred at the canopy top, while at the canopy bottom deposition was often three to four times lower. The range of deposition fluxes of NO_3^- and NH_4^+ were similar in each of the three seasons. A comparison of the results of this study with other sites in California indicated that deposition fluxes of the studied ions were elevated. In addition to the deposition of NO_3^- and NH_4^+ , conductance (K_1) of total NO_3^- , HNO_3 vapor, and particulate NH_4^+ to pine branch surfaces depended on the position in the canopy. The highest values were always measured at the canopy top.*

Introduction

Little is known about concentrations of nitrogenous (N) pollutants in forest locations in California (Bytnerowicz and Fenn 1996). Annular denuder systems (Possanzini and others 1983) allow for effective separation and precise determination of gaseous and particulate N pollutants. These systems have been successfully used in chaparral stands of the San Gabriel Mountains (Grosjean and Bytnerowicz 1993), mixed conifer forests in the central Sierra Nevada (Bytnerowicz and Riechers 1995) and coastal sage scrub communities in southern California (Allen and others, this volume). Short-term, preliminary measurements of N pollutants in mixed conifer forests in the San Bernardino Mountains have been reported (Fenn and Bytnerowicz 1993), but detailed information on their temporal and spatial distribution is lacking.

In the arid climates of the California mountains, dry deposition is one of the major mechanisms of ion transfer from the atmosphere to forests (Rundell and Parsons 1977). Foliage rinsing techniques have been successfully used for determinations of atmospheric deposition of various ions to trees (Bytnerowicz and Fenn 1996, Bytnerowicz and others 1987, Lindberg and others 1986). This technique is especially valuable in the dry climates of California where precipitation (which would allow for throughfall collection) is scarce in the summer season.

This paper presents results of a study that monitored concentrations of gaseous and particulate N air pollutants; measured deposition of nitrate (NO_3^-) and ammonium (NH_4^+) to branches of ponderosa pine seedlings; and calculated conductance to branch surfaces (K_1) of total nitrate, nitric acid vapor, and particulate ammonium on a vertical transect in a canopy of mature ponderosa/Jeffrey pines within the mixed conifer forest in the San Bernardino Mountains in southern California during the mid-May to mid-October periods of 1992 to 1994.

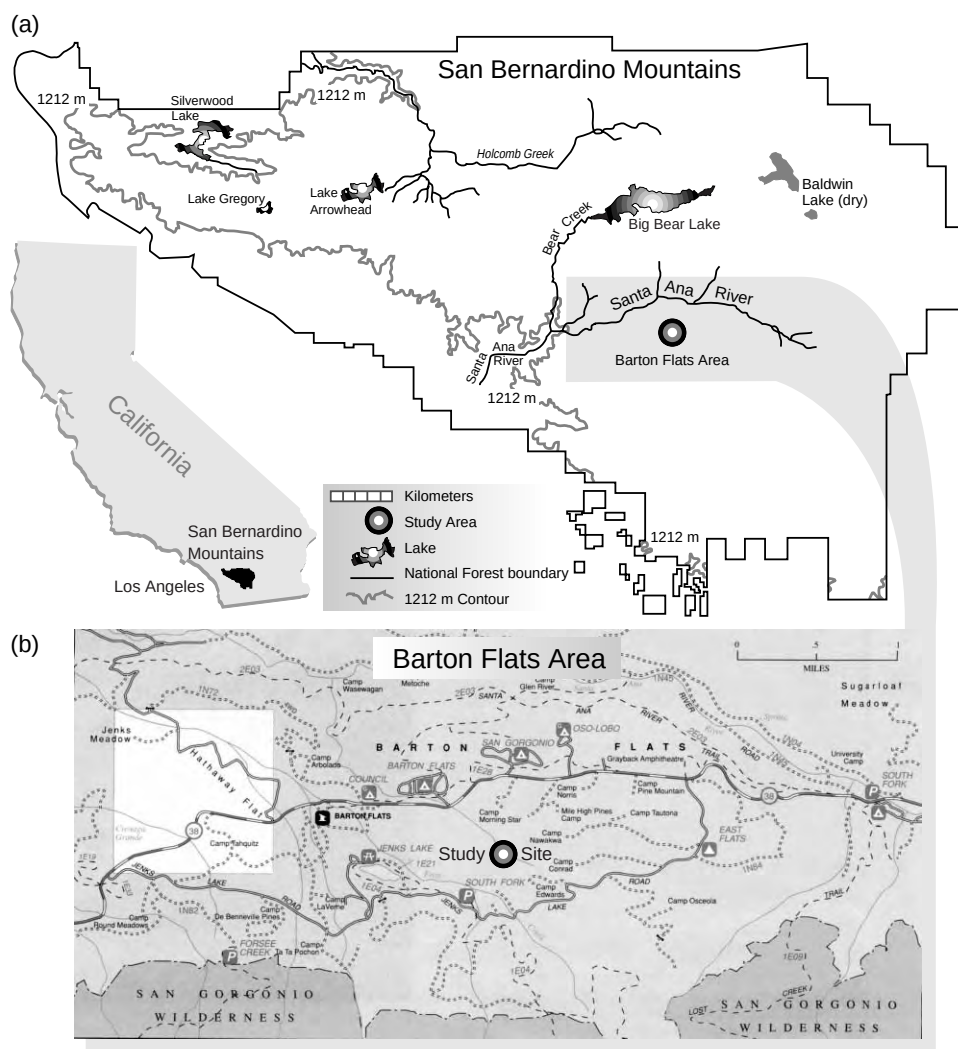
Methods

The study was performed in Barton Flats in the San Bernardino Mountains at an elevation of 2,100 m (fig. 1). A 30 m tall tower was located in the mixed conifer

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Figure 1 — The study site, Barton Flats, is located in the San Bernardino Mountains in southern California (a), south of State Highway 38 (b).



forest stand of mature ponderosa / Jeffrey pines (Arbaugh and others, this volume). The forest stand was open with about 67 percent of its ground area under tree canopy (Fenn and Bytnerowicz 1996). Single annular denuder systems were placed at each of four different levels of the tower: 29 m (canopy top), 24 m and 16 m (canopy middle), and 12 m (canopy bottom) (fig. 2). Average 24-hour concentrations of nitric acid (HNO_3) vapor, nitrous acid (HNO_2) vapor, ammonia (NH_3), particulate nitrate (NO_3^-), and particulate ammonium (NH_4^+) were determined with an annular denuder system (Peake and Legge 1987, Possanzini and others 1983). Constant air flow of 17 L min^{-1} through the systems was provided by steady flow pumps. This allowed for quantitative determinations of gaseous compounds deposited inside annular denuder tubes and the fine particle fraction ($< 2.2 \mu\text{m}$ in diameter) collected on teflon and nylon filters. Concentrations of NO_3^- and NH_4^+ in the annular tube extracts and filter extracts were determined with ion chromatography (Dionex 4000i chromatograph).³ Concentrations of N pollutants on the tower were measured six times during the 1992 summer season, eight times during the 1993 summer season, and four times during the 1994 summer season.

Deposition fluxes of NO_3^- and NH_4^+ to branches of ponderosa pine seedlings were determined on each of the four levels of the tower in the vicinity of the annular denuder systems. Four 2-year old seedlings were located at each of the tower levels. Seedlings were about 40-50 cm tall and were grown in 6 L pulp pots filled with 2:1:1 mixture of Oakley sand, peat, and fir bark. The seedlings were watered with tap water as needed (typically once a week). At the beginning of each collection

³ Mention of trade names or products is for information only and does not imply endorsement by the U.S. Department of Agriculture.

period, seedling branches about 10 cm long were thoroughly rinsed with double deionized water. The branches were re-rinsed at the end of 1-week long collection periods with 100 mL of double deionized water and the rinses were collected in 250 mL Nalgene bottles. Bottles were placed on ice, immediately transferred to the laboratory and placed at -18 °C. Concentrations of NO_3^- were determined with ion chromatography (Dionex 4000i ion chromatograph) and concentrations of NH_4^+ by colorimetry (TRAACS, Model 800 instrument) (Bytnerowicz and others 1987). Deposition was measured four times during the 1992 season, six times during the 1993 season, and five times during the 1994 photochemical smog season.

Conductances (K_i) of total NO_3^- ($K_{\text{NO}_3^-}$), HNO_3 vapor (K_{HNO_3}) and particulate NH_4^+ ($K_{\text{NH}_4^+}$) to surfaces of pine branches have been calculated (Bytnerowicz and others 1996). Conductance of these compounds is analogous to the deposition velocity at a canopy level (v_d) and is also expressed as cm s^{-1} (Hanson and Lindberg 1991). A general equation for conductance calculations is as follows:

$$K_i = F / c$$

in which:

F - deposition flux to branch surface ($\mu\text{g m}^{-2} \text{h}^{-1}$)

c - pollutant concentration ($\mu\text{g m}^{-3}$)

$K_{\text{NO}_3^-}$ was derived by dividing the NO_3^- deposition flux to branch surfaces by a sum of ambient concentrations of particulate NO_3^- and gaseous HNO_3 . K_{HNO_3} was calculated by dividing the deposition flux of NO_3^- to branches that could be attributed to HNO_3 by the ambient concentrations of HNO_3 . Deposition fluxes of NO_3^- attributed to HNO_3 were obtained by subtracting the portion of particulate NO_3^- deposition from the total NO_3^- deposition (calculated by multiplying ambient concentrations of particulate NO_3^- by the literature value of particulate NO_3^- deposition velocity to conifer foliage) (Davidson and Wu 1990). $K_{\text{NH}_4^+}$ was calculated by dividing NH_4^+ deposition to branches by the ambient concentrations of particulate NH_4^+ .

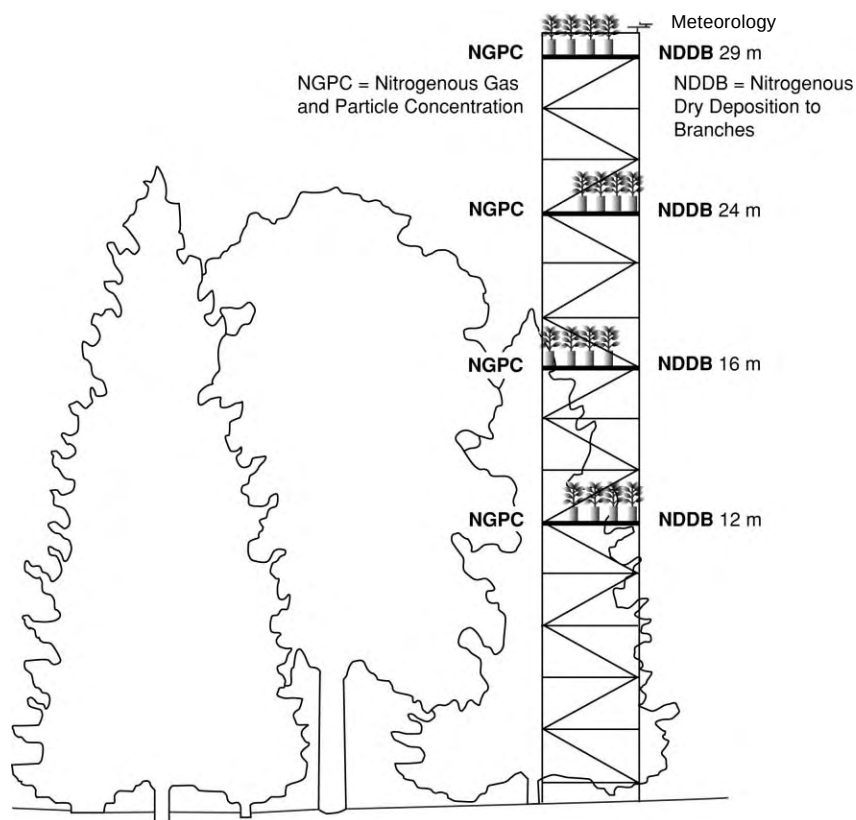


Figure 2 — Schematic of the tower in the mature ponderosa/jeffrey pine canopy indicating the four levels at which concentrations of N pollutants and their deposition to branches were measured.

Significance of the effects of position on the tower and a period of measurement on concentrations, deposition fluxes, and K_1 values of N pollutants during the 1992 to 1994 summer seasons were evaluated with a two-way analysis of variance and a Bonferroni t-test (SigmaStat 1994).

Results

Concentrations of N pollutants

Seasonal concentrations of HNO_3 vapor, NH_3 , particulate NO_3^- and NH_4^+ were averaged at different heights in the canopy for the 1992 to 1994 seasons (*table 1*). In the 1993 season the highest HNO_3 vapor concentrations occurred at 24 m and were significantly higher than the concentrations at 12 m. In 1992 and 1994, position within the canopy had no significant effect on HNO_3 concentrations. Position within the canopy did not affect the concentrations of NH_3 , NO_3^- , or NH_4^+ during the entire study.

During the 3 years of study, the highest concentration of HNO_3 vapor ($9.7 \mu\text{g m}^{-3}$) was measured in June 1994 and the lowest ($1.3 \mu\text{g m}^{-3}$) in May 1993. The highest concentration of NH_3 ($2.6 \mu\text{g m}^{-3}$) was recorded in July 1992, while the highest concentrations of particulate NO_3^- ($9.5 \mu\text{g m}^{-3}$) and particulate NH_4^+ ($2.0 \mu\text{g m}^{-3}$) were determined in June 1992. The lowest concentrations of NH_3 , particulate NO_3^- and particulate NH_4^+ were determined in September 1992 (1.0 , 0.35 , and $0.2 \mu\text{g m}^{-3}$, respectively). Concentrations of HNO_2 vapor during the 1992 season ranged from 0.10 to $0.42 \mu\text{g m}^{-3}$, from 0.00 to $0.35 \mu\text{g m}^{-3}$ in the 1993 season, and from 0.00 to $0.12 \mu\text{g m}^{-3}$ in the 1994 season. In general, the season significantly affected the concentrations of all studied N species, but no consistent trends were evident (Miller and others 1996).

Table 1 — Average 24-hour concentrations of N pollutants in the ponderosa/Jeffrey pine canopy during three summer seasons (1992-1994) of investigations ($\mu\text{g m}^{-3}$) Different letters assigned to the concentration numbers indicate statistically significant differences between the canopy levels.

Canopy level	1992	1993	1994
HNO_3			
29 m	3.65	3.24 ab	6.48
24 m	3.33	3.69 a	6.74
16 m	3.05	3.32 ab	6.78
12 m	3.00	3.14 b	5.86
P value	0.084	0.039	0.517
NH_3			
29 m	1.43	1.51	1.6
24 m	1.43	1.54	1.56
16 m	1.49	1.41	1.56
12 m	1.49	1.35	1.56
P value	0.841	0.119	0.963
NO_3^-			
29 m	3.86	2.37	2.33
24 m	3.3	2.44	2.33
16 m	3.32	2.72	2.37
12 m	2.97	2.92	1.91
P value	0.250	0.661	0.187
NH_4^+			
29 m	1.08	0.75	0.69
24 m	0.99	0.64	0.69
16 m	0.98	0.63	0.7
12 m	0.71	0.68	0.5

Surface Deposition of Washable Nitrate and Ammonium

Deposition of NO_3^- to branches of the ponderosa pine seedlings was significantly affected by position within the canopy (table 2). The highest deposition flux values were always determined at the canopy top (29 m) and approached about $200 \mu\text{g m}^{-2} \text{h}^{-1}$ during individual 1-week long determination periods (Bytnerowicz and others 1996). Deposition at 24 m was consistently lower than at the top but nearly always higher than at 16 m and the canopy bottom (12 m). The highest deposition fluxes occurred in July and August in each of the three years. Nitrate deposition values were similar during the three years (Bytnerowicz and others 1996).

Ammonium deposition was also strongly affected by position within the canopy (table 2). The highest deposition flux values occurred at the canopy top and were sometimes as high as $20 \mu\text{g m}^{-2} \text{h}^{-1}$ during individual determination periods (Bytnerowicz and others 1996). Deposition at 24 m was higher than at 16 m and at 12 m. Although significant differences in deposition fluxes between the individual collection periods occurred, no clear seasonal trend was observed. The values of NH_4^+ deposition flux were similar in all three seasons (Bytnerowicz and others 1996).

Table 2—Average seasonal deposition fluxes of NO_3^- and NH_4^+ ($\mu\text{g m}^{-2} \text{h}^{-1}$) to seedling branches within the ponderosa/Jeffrey pine canopy during three summer seasons (1992-1994). Different letters assigned to the deposition numbers indicate statistically significant differences between the canopy levels.

Canopy level	1992	1993	1994
NO_3^-			
29 m	126.2 a	118.6 a	132.6 a
24 m	67.6 b	79.8 b	84.0 b
16 m	40.7 c	51.9 c	62.9 bc
12 m	35.4 c	52.4 c	54.7 c
P value	<0.0001	<0.0001	<0.0001
NH_4^+			
29 m	13.29 a	14.57 a	12.00 a
24 m	9.77 b	10.01 b	9.02 b
16 m	6.61 c	8.13 c	8.28 b
12 m	6.33 c	7.02 c	5.74 c
P value	<0.0001	<0.0001	<0.0001

NO_3^- , HNO_3 and NH_4^+ Conductance at a Branch Level

The highest conductance (K_i) values for total NO_3^- , HNO_3 vapor, and particulate NH_4^+ (table 3) were recorded at the canopy top and the lowest at the bottom of the canopy. For total NO_3^- and HNO_3 vapor these differences were strongly significant during three seasons. For particulate NH_4^+ a significant effect of the position within the canopy was seen only in 1993. Large differences were found between individual periods of measurements within each season, i.e., during the 1993 season the range of calculated K_{HNO_3} values for the canopy top was between 0.4 cm s^{-1} and 3.1 cm s^{-1} (Bytnerowicz and others 1996). Significant differences between the $K_{\text{NO}_3 \text{ total}}$ and K_{HNO_3} values during individual collection periods occurred in 1993 and 1994, and between the K_{NH_4} values only in 1993.

Discussion

Position within the canopy did not affect concentrations of the studied pollutants probably because of the openness of the canopy upwind from the scaffold (with

Table 3—Average conductance values (K_i) of N species to single ponderosa pine branches during 3 years (1992-1994) of investigations (cm s^{-1}). Different letters assigned to the K_i values indicate statistically significant differences between canopy levels.

Canopy level	1992	1993	1994
NO_3^- total			
29 m	0.50 a	0.61 a	0.42 a
24 m	0.31 ab	0.45 ab	0.27 ab
16 m	0.16 b	0.29 bc	0.21 b
12 m	0.16 b	0.26 c	0.19 b
P value	<0.002	<0.000	<0.002
HNO_3			
29 m	0.83 a	1.26 a	0.57 a
24 m	0.48 ab	0.74 ab	0.36 ab
16 m	0.30 b	0.55 b	0.26 b
12 m	0.25 b	0.52 b	0.26 b
P value	<0.010	<0.003	<0.011
NH_4^+			
29 m	0.28	0.66a	0.66
24 m	0.22	0.50 ab	0.29
16 m	0.16	0.40 b	0.25
12 m	0.18	0.34 b	0.29
P value	0.099	0.012	0.160

the exception of significantly higher concentrations of HNO_3 at 24 m than at 12 m during the 1993 season). In the westerly direction, from which pollutants were transported, no nearby canopy inhibited vertical mixing. In more dense forest canopies, such as in the eastern United States, a vertical gradient of pollutants with diminishing concentrations from canopy top to bottom is typical (Meyers and others 1989).

Concentrations of HNO_3 vapor at Barton Flats were moderately high (seasonal means 3.0 to 6.8 $\mu\text{g m}^{-3}$) compared to the summer average concentrations in forest locations in Tennessee (3.0 $\mu\text{g m}^{-3}$) and near Gottingen, Germany (5.8 $\mu\text{g m}^{-3}$) (Lindberg and others 1990). At Tanbark Flat in the San Gabriel Mountains of southern California, a mountain site which is considered one of the most polluted in the United States (Bytnerowicz and others 1987), the average 24-hour concentrations during the 1988 to 1991 summer seasons ranged from 6.7 to 11.7 $\mu\text{g m}^{-3}$ (Grosjean and Bytnerowicz 1993). At various locations in the Sierra Nevada the average 24-hour summer concentrations were below 1 $\mu\text{g m}^{-3}$ (Ashbaugh and others 1991).

Nitrous acid vapor concentrations (seasonal means 0.00 to 0.42 $\mu\text{g m}^{-3}$) were lower than the summer average concentrations at the Tanbark Flat site (0.55 to 0.65 $\mu\text{g m}^{-3}$) (Grosjean and Bytnerowicz 1993). However, they were higher than the annual mean concentration of 0.05 $\mu\text{g m}^{-3}$ reported at the remote location in the Canadian Rockies (Legge and Krupa 1989).

Ammonia concentrations (seasonal means 1.35 to 1.6 $\mu\text{g m}^{-3}$) were elevated and similar to the seasonal means 1.1 to 1.85 $\mu\text{g m}^{-3}$ determined at Tanbark Flat (Grosjean and Bytnerowicz 1993) and lower than the estimated seasonal mean 3.0 $\mu\text{g m}^{-3}$ determined near Gottingen, Germany (Lindberg and others 1990). The measured concentrations were several times higher than the annual mean 0.26 $\mu\text{g m}^{-3}$ value measured in the Canadian Rockies (Legge and Krupa 1989).

and the estimated seasonal mean $0.2 \mu\text{g m}^{-3}$ in Tennessee (Lindberg and others 1990). However, concentrations of NH_3 in the Netherlands and Great Britain may be several times higher than the reported values reaching $15 \mu\text{g m}^{-3}$ (Draaijers and others 1989, Keuken and others 1988).

Concentrations of particulate NO_3^- (seasonal means 1.9 to $3.9 \mu\text{g m}^{-3}$) were high and similar to the values determined at Tanbark Flat (seasonal means 1.75 to $2.9 \mu\text{g m}^{-3}$) (Grosjean and Bytnerowicz 1993). These values greatly exceeded the annual mean $0.13 \mu\text{g m}^{-3}$ reported for the Canadian Rockies (Legge and Krupa 1989), and seasonal mean $0.19 \mu\text{g m}^{-3}$ determined in Tennessee (Lindberg and others 1990). The reported values were similar to 2.1 to $5.1 \mu\text{g m}^{-3}$ measured in the San Bernardino Mountains (Fenn and Bytnerowicz 1993) and $3.3 \mu\text{g m}^{-3}$ measured in the Los Angeles Basin, California (Sickles and others 1988). Mean NO_3^- concentration in Claremont, Los Angeles Basin ($7.9 \mu\text{g m}^{-3}$) (Pierson and Brachaczek 1988) was higher than the concentrations determined in this study.

Concentrations of particulate NH_4^+ (seasonal means 0.5 to $1.1 \mu\text{g m}^{-3}$) were similar to the values determined at Tanbark Flat (seasonal means 0.65 to $0.85 \mu\text{g m}^{-3}$) (Grosjean and Bytnerowicz 1993) and other locations in the San Bernardino Mountains (0.1 to $0.9 \mu\text{g m}^{-3}$) (Fenn and Bytnerowicz 1993). Ammonium concentrations were also similar to the mean concentration of $1.0 \mu\text{g m}^{-3}$ determined in the urban Los Angeles area (Sickles and others 1988). Ammonium concentrations reported for the forest locations in Tennessee and Germany (seasonal means 2.4 and $5.5 \mu\text{g m}^{-3}$, respectively) (Lindberg and others 1990) were higher than in this study.

Nitrate fluxes to branches were elevated; the highest values during individual collection periods (approaching $200 \mu\text{g m}^{-2} \text{h}^{-1}$) were similar to the value of $180 \mu\text{g m}^{-2} \text{h}^{-1}$ determined for mature ponderosa pine in Camp Paivika of the San Bernardino National Forest (Fenn and Bytnerowicz 1993). Higher NO_3^- deposition values were determined at Tanbark Flat; deposition to hoaryleaf ceanothus (*Ceanothus crassifolius*) ranged from 88 to $455 \mu\text{g m}^{-2} \text{h}^{-1}$ (Bytnerowicz and others 1987). Deposition of nitrate to native pines in other parts of the state were much lower: about 4 to $6 \mu\text{g m}^{-2} \text{h}^{-1}$ for Eastern Brook Lake in the eastern Sierra Nevada (Bytnerowicz and others 1992); 27 to $30 \mu\text{g m}^{-2} \text{h}^{-1}$ for Emerald Lake in the western Sierra Nevada (Bytnerowicz and others 1991); and 6 to $31 \mu\text{g m}^{-2} \text{h}^{-1}$ for Whitaker Forest in the western Sierra Nevada (Bytnerowicz and Riechers 1995).

Deposition fluxes of NH_4^+ were also elevated at Barton Flats; the highest values determined during individual collection periods approached $20 \mu\text{g m}^{-2} \text{h}^{-1}$ (Bytnerowicz and others 1996). These values were similar to deposition of $21 \mu\text{g m}^{-2} \text{h}^{-1}$ determined at Camp Paivika, a high pollution site in the western San Bernardino Mountains (Fenn and Bytnerowicz 1993). Deposition of NH_4^+ to hoaryleaf ceanothus at Tanbark Flat (17 to $46 \mu\text{g m}^{-2} \text{h}^{-1}$) was higher than at Barton Flats (Bytnerowicz and others 1987). Deposition to native pines at some other forest sites in California were lower than in the present study: at Eastern Brook Lake ranged from 0.8 to $1.6 \mu\text{g m}^{-2} \text{h}^{-1}$ (Bytnerowicz and others 1992); Emerald Lake, 6.0 to $9.0 \mu\text{g m}^{-2} \text{h}^{-1}$ (Bytnerowicz and others 1991); and Whitaker Forest, 1.0 to $1.5 \mu\text{g m}^{-2} \text{h}^{-1}$ (Bytnerowicz and Riechers 1995). The results of this study have clearly indicated that NO_3^- and NH_4^+ deposition fluxes in a mixed coniferous forest strongly depend on the position of the branches within the canopy. Ion deposition increased with increasing branch height. Because concentrations of gaseous and particulate pollutants did not depend on canopy position, the observed distribution of deposition fluxes within the forest canopy could not be related to the air pollution concentration differences. Difference in deposition fluxes could be attributed to varying wind speed occurring along the vertical transect within the canopy because the highest deposition values occurred at the canopy top where wind speeds were highest (Arbaugh and others, this volume). The HNO_3 conductance (k_{HNO_3}) values at the canopy top reached 3.1 cm s^{-1} (Bytnerowicz and others 1996) and were similar to the k_{HNO_3} values of 0.6 to 3.4 cm s^{-1} determined for loblolly pine in Tennessee (Hanson and Garten 1992). In general, the conductance of a pollutant to a branch

multiplied by the ambient pollutant concentration provides an estimate of deposition at the branch level. Branch level deposition multiplied by the canopy leaf area index (LAI) provides an estimate of canopy level deposition. Therefore, theoretically, the K_{HNO_3} and ambient HNO_3 concentration values can be used for an estimate of surface NO_3^- deposition to ponderosa pine canopies from atmospheric HNO_3 . Similarly, information on conductance to branches of total NO_3^- and particulate NH_4^+ can be used for estimates of surface dry deposition of total NO_3^- and NH_4^+ if information on ambient concentrations of HNO_3 vapor + particulate NO_3^- or particulate NH_4^+ are provided. On the basis of the results of this study, and other related companion studies, empirical models for estimating N atmospheric deposition to the mixed conifer forest stands in California are being developed (Arbaugh and others, this volume).

Total N deposition to the forest stand in Barton Flats was estimated based on the presented results, information on deposition to other dominant tree species, and LAI for the overstory species (Bytnerowicz and others 1996). The estimated forest stand deposition for the 1993 season was 5 to 9 kg ha⁻¹ yr⁻¹, and occurred mostly as dry deposition to surfaces of plants and soil (Bytnerowicz and others 1996). Judging from the results of this and other studies (Fenn and Bytnerowicz 1993, Grosjean and Bytnerowicz 1993), Barton Flats can be characterized as a site with elevated N deposition located in the middle to low end of the west to east air pollution gradient in the San Bernardino Mountains. Chronic N deposition in the more polluted regions of the San Bernardino and San Gabriel Mountains is associated with symptoms of N excess as described by Aber and others (1989), which are especially evident as high NO_3^- concentrations in soil solution and streamwater (Fenn and others 1996, Riggan and others 1985). Long-term effects of N deposition on plant community dynamics in these forests are as yet unknown.

In summary, concentrations of the measured N species at the Barton Flats site were elevated compared to the pristine mountain areas in California and elsewhere. No differences in concentrations of the measured N species were found on the vertical transect in the mature pine canopy. However, deposition fluxes of NO_3^- and NH_4^+ as well as conductance (K_1) of gaseous HNO_3 , total NO_3^- , and particulate NH_4^+ were always the highest at the canopy top.

Acknowledgments

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