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Determination of Acidic Gas and Particle Concentrations in Open-Top Field Chambers

CALIFORNIA ENVIRONMENTAL PROTECTION AGENCY

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Determination of Acidic Gas and Particle Concentrations in Open-Top Field Chambers

Final Report

to the

California Air Resources Board

for

Contract No. A132-174

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Abstract

This report describes determinations of concentrations of gaseous and particulate air pollutants in open-top field chambers used for exposing of tree seedlings to acidic rain and ozone. The study was performed at the Chico Tree Improvement Facility, Chico, California, in summer 1992. Measurements were performed in standard chambers used for air pollution exposures. During the study, air pollution concentrations were measured in the following types of chambers: charcoal-filtered (control); ambient air with ambient ozone concentrations; ambient air with double ambient ozone concentrations. In addition, measurements were also performed in outside, chamberless plots. Concentrations of gaseous nitric acid, gaseous nitrous acid, ammonia, sulfur dioxide and nitrate, ammonium and sulfate in fine and coarse particulate fractions were determined. Increased concentrations of nitric acid were detected in chambers due to the generation of ozone, however, these concentrations were lower than the ambient concentrations measured in outside plots. Elevated concentrations of nitrous acid were detected in the charcoal-filtered chambers. Nitrous acid was probably produced during the heterogenous reactions of NO and NO_2, including hydrolysis of NO_2 occurring on charcoal filters. Concentrations of nitric acid and nitrous acid determined in this study are compared to concentrations from other locations in North America and Europe.

Acknowledgements

The author would like to thank Mr. Minh Tran for his excellent technical assistance during the field study. Ms. Kathy Meade, Ms. Carol Kenefick and other technical personnel of the chemical laboratory, USDA Forest Service Fire Laboratory in Riverside, performed chemical analysis of the samples. Dr. Paul Anderson provided assistance during the field experiment and gave the necessary technical information and ozone data. Dr. Brent Takemoto helped in coordinating this study and provided editorial advice during preparation of this report.

Disclaimer

The statements and conclusions in this report are those of the contractor and not necessarily those of the California Air Resources Board. The mention of commercial products, their source, or their use in connection with material reported herein is not to be construed as either an actual or implied endorsement of such products.

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Summary and Conclusions

Air pollutants, especially ozone, may be highly toxic to California forests (Miller and Millecan, 1971). However, very little is known about the effects of other air pollutants on forests in this part of the United States. In our previous studies, we found that fluxes of nitrate, sulfate, ammonium, hydrogen ion, and metallic cations to native pines in the western Sierra Nevada were much higher than fluxes in the eastern Sierras (Bytnerowicz et al., 1991, 1992). For the nitrate ion, increased deposition is in part the result of gaseous nitric acid deposition, a compound which is found in elevated concentrations in the western Sierras (Bytnerowicz and Miller, 1991).

Open-top chambers developed by Heagle et al. (1973) have been widely used to study the effects of air pollution on plants (including tree seedlings). For many years, little attention has been paid to alterations of the chemical environment of the chambers caused by the filtration of air through particulate filters or charcoal filters. It has been demonstrated that various particulate filters reduce concentrations and depositions of nitrate, ammonium, sulfate and other ions compared to ambient air. It has also been shown that while charcoal filtration reduces concentrations of ozone in the chambers, it increases concentrations of nitric oxide (Bytnerowicz et al., 1989). In addition to these changes in the environment of the chambers, concerns have been raised that the production of ozone by ozone generators using high voltage electrical discharge and dry ambient air (instead of oxygen), can produce elevated concentrations of gaseous nitric acid (Brown and Roberts, 1988).

The present study was performed in order to determine concentrations of gaseous and particulate nitrogen and sulfur air pollutants in open-top chambers and chamberless, outside plots used in a study to determine the effects of acidic rain and ozone on ponderosa pine. Concentrations of gaseous nitric acid, gaseous nitrous acid, ammonia, sulfur dioxide as well as nitrate, ammonium and sulfate in fine and coarse particulate fractions were determined during a 48 hr intensive study performed in summer 1992. The nitric and nitrous acid measurements were compared with results from other locations in North America and Europe.

The following conclusions have been drawn from the study:

1. Gaseous nitric acid was produced during the generation of ozone from ambient air, however, concentrations of this gas in the chambers were lower than in ambient air.

2. Nitrous acid was produced in the charcoal-filtered chambers due to the heterogenous reactions of NO and NO_2, including hydrolysis of NO_2 occurring on charcoal filters, and possibly reaction of NO with $^{\cdot}OH$ radicals. The nighttime concentrations of the pollutant were much higher than daytime levels.

3. As expected, concentrations of sulfur dioxide in the charcoal-filtered chambers were much lower than in the ambient chambers and the outside plots.

4. The filtration of air through charcoal reduced nitrate concentrations in the coarse particulate fraction, but no other effects on concentrations of ions in the fine or coarse particulate fractions were observed.

Recommendations

1. Much more attention should be given to the evaluation of the chemical environment of chambers used for exposing plants to air pollutants.

2. Pilot studies aimed at measuring the products of the air filtration and pollutant generating systems (e.g. ozone) should always be performed.

3. Careful examination of the results of plant exposures, especially from long-term studies, is needed. Special attention should be paid to the unexpectedly poor performance of plants in charcoal-filtered (control) chambers. This is especially important for past and present studies done in areas with high ambient concentrations of nitric acid and nitrogen dioxide. In such conditions, the presence of elevated concentrations of potentially toxic nitrous acid and nitric oxide may be expected.

4. For long-term studies, ozone should be generated from oxygen in order to avoid increased concentrations of nitric acid. Although the resulted nitric acid concentrations generated in the present study were too low to cause acute toxic effects on plants, increased levels of airborne nitrogen could provide additional fertilizer to the plants.

I. INTRODUCTION

The trees in the western Sierra Nevada, California, have long been known to be damaged by elevated concentrations of photochemical pollutants from polluted air masses originating in the California Central Valley (Duriscoe and Stolte, 1989; Peterson et al., 1987). However, very little is known about the effects of other pollutants on forest vegetation in the mountains of California.

Forests in the western United States, and especially in California, are exposed to wet and dry deposition throughout the year. Inputs of acidity during the winter season are mainly provided by snow, rain, fog, and clouds with little contribution from dry deposition. During the rest of the year, dry deposition of gases and particles dominates inputs of acidity to forests. Dry deposition of nitrogenous compounds seems to be of special importance in the western United States (Young et al., 1988). Elevated concentrations of gaseous nitrous and nitric acids were found in selected California mountain sites (Bytnerowicz et al., 1987; Bytnerowicz and Miller, 1991; Fenn and Bytnerowicz, 1992). High concentrations of the gaseous and particulate nitrogenous pollutants in source areas (Los Angeles Basin and San Joaquin Valley) caused increased depositions of the nitrogenous and sulfur compounds to forest vegetation in the surrounding mountains. Deposition of nitrate, ammonium and sulfate to the foliage of native pines of the subalpine zone of the western Sierra Nevada were much higher than on the eastern side of this mountain range (Bytnerowicz et al., 1991; 1992). Exposures of plants to elevated concentrations of nitrogenous compounds may have direct toxic effects (e.g., gaseous nitric acid on plant cuticles) and also may affect the nutritional status of forest vegetation. It is believed that dry deposition of gases and particles may substantially contribute to the nutrient pool in forest ecosystems, and therefore may have important long-term effects (Waring and Schlesinger, 1985). These effects may be initially positive (increased deposition of N compounds to nutrient-poor forest sites - fertilization), however, the increased nitrogen deposition over the long-term may cause acidification of soils and nutritional imbalances (deficiencies of macro- and micronutrients).

In studies on air pollution effects on plants, the open-top chambers developed by Heagle et al. (1973) have been widely used. The purpose of using the chambers was to compare the performance of plants in polluted air with the plants exposed to the air from which pollutants of interest (mainly SO_2 or O_3) have been removed - (Heagle and Philbeck, 1979). However, relatively little is known about how the air filtration and ozone production processes affect the chemical environment of the chambers. It was shown that filtration significantly reduced concentrations of airborne ions in open-top chambers, and that the passage of air through activated charcoal causes a chemical reduction of nitrogen dioxide leading to elevated concentrations of nitric oxide inside the chambers (Bytnerowicz et al., 1989). In addition to these problems, in ozone exposures studies, there is the possibility of contaminating the chamber air with nitric acid during ozone

generation from ambient air (Brown and Roberts, 1988).

The present study which was sponsored in part by the California Air Resources Board, has been done in cooperation with researchers from the University of California, Berkeley. The purpose of the study was to evaluate the chemical environment in open-top chambers used for exposing ponderosa pine seedlings to acidic rain and ozone.

II. PROJECT OBJECTIVE

The objective of this project was to measure daytime and nighttime concentrations of gases and particles in open-top field chambers used to determine the effects of acidic rain and ozone on ponderosa pine seedlings (Contract No. A132-101; University of California, Berkeley). The data will be used to estimate annual fluxes of acidic inputs (i.e. wet plus dry deposition) to the trees in the acidic rain and ozone effects study.

III. MATERIALS AND METHODS

Concentrations of gaseous nitric acid, nitrous acid, ammonia and sulfur dioxide as well as concentrations of nitrate, ammonium and sulfate ions in fine particles (<2.2 um in diameter) and coarse particles (>2.2 um) were determined with annular denuder systems developed by Possanzini et al. (1983) and modified by Peake and Legge (1987). The annular denuder systems were placed inside open-top chambers (Heagle et al., 1973) at the Chico Tree Improvement Facility, Chico, California. The following filtration regimes were examined:

(a) charcoal-filtered air (two stage particulate filter consisting of a coarse fiberglass layer coupled with a fine fiberglass strainer mat [Grillo's Filters, Riverside, CA] and and an activated charcoal filter [Barnebey and Cheney, Columbus, OH]).

(b) ambient air (two stage particulate filter [Grillo's Filters, Riverside, CA]).

(c) ambient air with O_3 added at 2 x ambient concentrations (two stage particulate filter [Grillo's Filters, Riverside, CA]).

The fourth annular denuder system was placed in an outside plot to determine ambient concentrations of the pollutants.

Ozone was generated from compressed ambient air that had been passed through a desiccation system consisting of a condenser, two water traps and four chemical desiccant columns. The dry air was passed through an ozone generator (Model GL-1, PCI Inc., NJ) converting ambient oxygen to ozone by means of high voltage electrical discharge. Air flow was maintained at 30 L/ min with a line pressure of 15 psi. The ozonated air stream was directed through individual flowmeters to the exposure chambers through Teflon tubing. Ozone concentrations were monitored with a Dasibi 1003 AH and the results were provided by Dr. Paul Anderson, University of California, Berkeley.

The study was performed on July 29-31, 1992, and the air samples were collected during two daytime (0600-1700 PST) and two nighttime (1800-0500 PST) periods. Extractions of the annular denuder tubes and filters, as well as the chemical analysis (ion chromatography with a Dionex 4000i for anions and colorimetry with a TRAACS 800 for ammonium) were performed at the USDA Forest Service, Pacific Southwest Research Station in Riverside.

IV. RESULTS AND DISCUSSION

Both the averages and ranges of ozone concentrations were similar for the day- and nighttime measurements (Table 1). This was due to the late occurrence of ozone peak values (highest values recorded about 1900-2000 PST). Concentrations of ozone in the ambient O_3 chambers were similar to the outside plots. Concentrations of ozone in the 2 x O_3 chambers were the highest, but always lower than the target levels. Filtration of air through charcoal (CF chambers) reduced the maximum ozone concentrations to the values below 31 ppb.

No major differences in concentrations of nitrate, sulfate and ammonium in the fine particulate fraction were observed among the chambers and between the chambers and the outside plot (Table 2). Similarly, no clear effects of air filtration on concentrations of sulfate in the coarse particles were seen, but concentrations of nitrate in the charcoal-filtered chambers were the lowest (Table 3). The ammonium ion levels in the coarse particulate fraction were below the detection limit.

Clear and consistent differences in concentrations of gaseous nitric and nitrous acids have been determined. During all of the collection periods concentrations of nitric acid were the highest in the outside plot, followed by the 2 x O_3, ambient O_3 and the CF chambers (Figure 1). This indicates that nitric acid was produced during ozone generation, however, in concentrations lower than in ambient air. No phytotoxic effects of nitric acid at these concentrations are expected, however, increased fertilization of trees with nitrogen could be anticipated. This is due to a very high deposition velocity (up to 2.25 cm/ sec) for gaseous nitric acid (Murphy and Sigmon, 1990). Long-term exposures to even slightly elevated concentrations of this gas may cause much higher than normal amounts of dry-deposited nitrogen to plants.

Table 1. Concentrations of ozone in open-top chambers and outside plots (ppb).

Date	CF chambers	1 x O_3 chambers	2 x O_3 chambers	Outside plots
July 29, 1992 day (1300-1700 PST)[1]	25 (22-31)[2]	60 (58-61)	105 (101-107)	70 (67-71)
July 29/30, 1992 night (1800-0500 PST)	18 (10-29)	45 (17-71)	60 (17-122)	46 (14-80)
July 30, 1992 day (0600-1700 PST)	17 (11-27)	38 (16-60)	66 (17-105)	42 (12-69)
July 30/31, 1992 night (1800-0500 PST)	19 (8-29)	41 (13-71)	62 (15-118)	41 (12-72)

[1]due to technical problems ozone data available only for a portion of a day.

[2]average concentrations with range of concentrations in parentheses.

Table 2. Concentrations of ions in fine particles (<2.2 micrometer diameter) in open-top chambers and outside plots (microgram/ m^3).

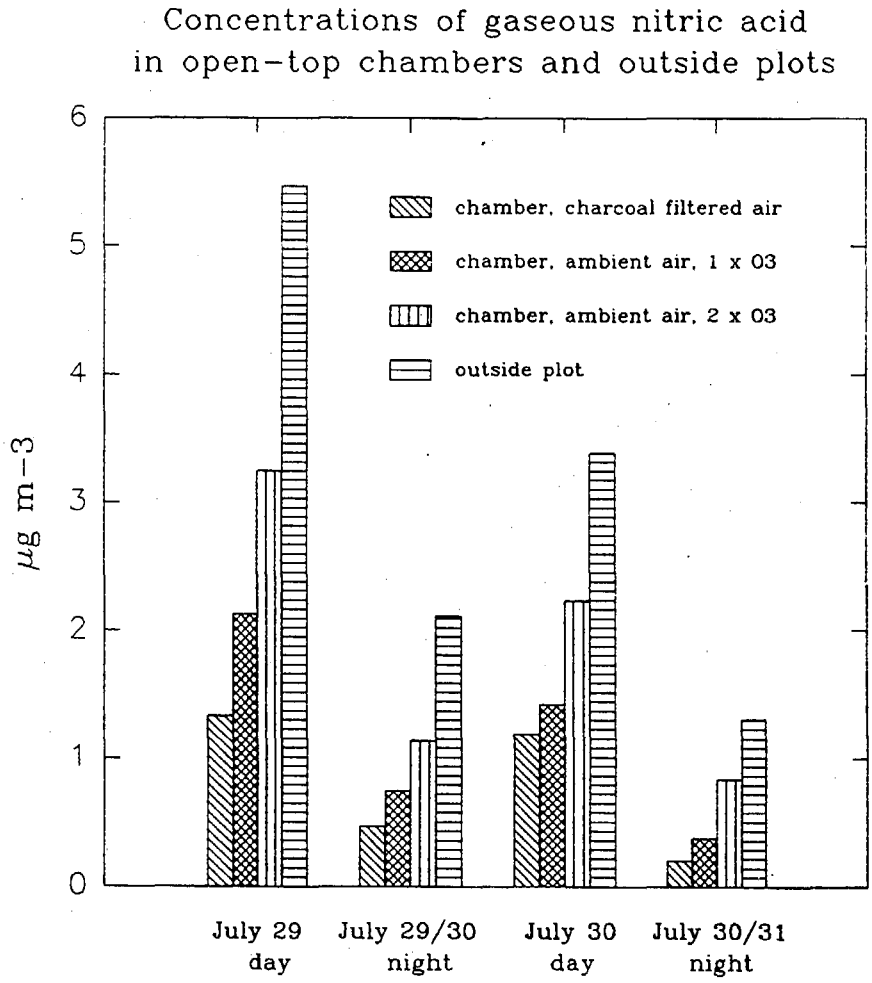
Date	CF chambers	1 x O_3 chambers	2 x O_3 chambers	Outside plots
Nitrate				
July 29, 1992 day	0.870	0.821	0.652	0.749
July 29/30 night	0.334	0.600	-[1]	0.325
July 30, 1992 day	0.135	0.000	0.270	0.337
July 30/31 night	0.000	0.131	0.044	0.066
Sulfate				
July 29 day	2.015	3.929	2.470	1.687
July 29/30 night	2.228	2.955	-[1]	1.629
July 30 day	0.306	0.576	0.171	0.481
July 30/31 night	0.083	0.074	0.070	0.387
Ammonium				
July 29 day	0.420	0.000	0.241	0.193
July 29/30 night	0.222	0.132	-[1]	0.489
July 30 day	0.090	0.180	0.045	0.090
July 30/31 night	0.044	0.044	0.044	0.088

[1]results not presented - contaminated samples.

Table 3. Concentrations of ions in coarse particles (>2.2 micrometer diameter) in open-top chambers and outside plots (microgram/ m^3).

Date	CF chambers	1 x O_3 chambers	2 x O_3 chambers	Outside plots
Nitrate				
July 29 day	0.120	0.722	0.434	0.967
July 29/ 30 night	0.089	0.178	0.311	0.489
July 30 day	0.050	0.315	0.050	1.170
July 30/ 31 night	0.000	0.087	0.305	0.304
Sulfate				
July 29 day	0.060	0.854	0.048	0.242
July 29/ 30 night	0.000	0.133	0.000	0.044
July 30 day	0.000	0.315	0.000	0.945
July 30/ 31 night	0.000	0.000	0.000	0.000

Figure 1


Concentrations of gaseous nitric acid
in open-top chambers and outside plots
μg m-3
0
1
2
3
4
5
6
chamber, charcoal filtered air
chamber, ambient air, 1 x O3
chamber, ambient air, 2 x O3
outside plot
July 29
day
July 29/30
night
July 30
day
July 30/31
night

The data collected in this study was compared with the results of measurements of ambient concentrations of nitric acid in other locations (Table 4). The comparison indicates that ambient concentrations of nitric acid at the Chico site were high, exceeding the ranges of concentrations determined in western Sierra Nevada locations (Whitaker Forest and Shirley Meadow). However, these concentrations were lower than at Tanbark Flat, San Gabriel Mountains, and the high pollution site Camp Paivika, San Bernardino Mountains. The measured concentrations were much higher than the concentrations at forest sites in other parts of the country.

Concentrations of gaseous nitrous acid in the ambient chambers and outside plots were similar. However, in the CF chambers, the concentrations of nitrous acid were increased and several times higher than in other chambers or the outside plot. Higher nighttime concentrations of nitrous acid in the CF chambers co-occurred with the elevated ambient concentrations (Figure 2). Our findings suggest that this increase of HNO_2 concentrations was caused by the heterogenous reactions of NO and NO_2, including hydrolysis of NO_2 occurring on charcoal filters (Dr. James Pitts, personal communication). A possibility of a reaction of NO with OH radical leading to elevated levels of HNO_2 should be also considered (Vecera and Dasgupta, 1991), however, such reaction is unlikely at night when the levels of HNO_2 increased the most (Dr. James Pitts and Dr. Roger Atkinson, personal communications). The daytime HNO_2 concentrations were lower than the nightime values due to its photolysis (Cox, 1974). Very little is known about the uptake of nitrous acid by plants or its phytotoxic effects. It is expected, however, that nitrite levels in plants could be elevated due to exposures to nitrous acid. According to Wellburn (1990), nitrite may be extremely toxic to plants.

Results of nitrous acid measurements in other locations are presented in Table 5. The ambient concentrations of nitrous acid at the Chico site were in the range of concentrations determined at Shirley Meadow, in the southwestern Sierra Nevada, but much higher than at other mountain locations (except Tanbark Flat of the San Gabriel Mountains). The measured concentrations of nitrous acid in the charcoal- filtered (control) chambers were similar to the values determined in urban locations.

Concentrations of ammonia were similar in all types of chambers and did not differ from the levels in the outside, chamberless plot (Figure 3). Concentrations of sulfur dioxide in the CF chamber were much lower than in the other chambers and the outside plot (Figure 4).

Clearly more investigations of the effects of various air filtration regimes as well as ozone production on the chemical environment of the exposure chambers is needed. Special attention should be given to possible toxic and fertilizing effects of the products of these processes.

Table 4. Concentrations of gaseous nitric acid in various locations (microgram/ m^3).

Location	Period and time of measurements	Range	Mean	Reference
Fortress Mt., Rocky Mtns, Alberta	3 yrs, 24 h averages	0.0005-3.50	0.31	Legge and Krupa, 1989
Niwot Ridge, Rocky Mtns, CO	October - November	0.03-0.48	0.14	Roberts et al., 1988
Point Arena, Pacific Coast, CA	May, 24 h averages	0.11-0.80	0.25	Roberts et al., 1988
Eastern Brook Lake, Sierra Nevada, CA	Winter, 24 h Summer, 24 h		0.15 0.37	Miller and Walsh, 1991
Smoky Mtns, TN	August, day Sept., night	0.09-1.00 0.04-0.20	0.54 0.12	Cadle and Mulawa, 1988
Whitaker Forest, Sierra Nevada, CA	Summer, day " night	0.90-3.80 0.10-1.10	1.83 0.59	Temple et al., 1990
Shirley Meadow, Sierra Nevada, CA	Summer, day " night	2.30-2.80 1.00-2.20	2.60 1.46	Temple and Bytnerowicz 1991
Camp Osceola, San Bernardino Mtns, CA	Sept., 24 h		1.10	Fenn and Bytnerowicz 1992
Camp Paivika, San Bernardino Mtns, CA	Sept., 24 h		8.90	Fenn and Bytnerowicz 1992
Tanbark Flat, San Gabriel Mtns, CA	Summer, day " night	15.40-30.20 0.90- 4.50	24.60 2.89	Grosjean and Bytnerowicz 1993
Claremont, Los Angeles Basin, CA	Summer	0.20-35.00		Appel et al., 1988
Warren, Detroit area, MI	Summer, 24 h Fall, 24 h Winter, 24 h Spring, 24 h	0.60-4.00 0.50-2.00 0.20-4.50 0.40-2.00		Cadle, 1985
Chico, CA	July, day " night		4.43 1.71	this study

Figure 2

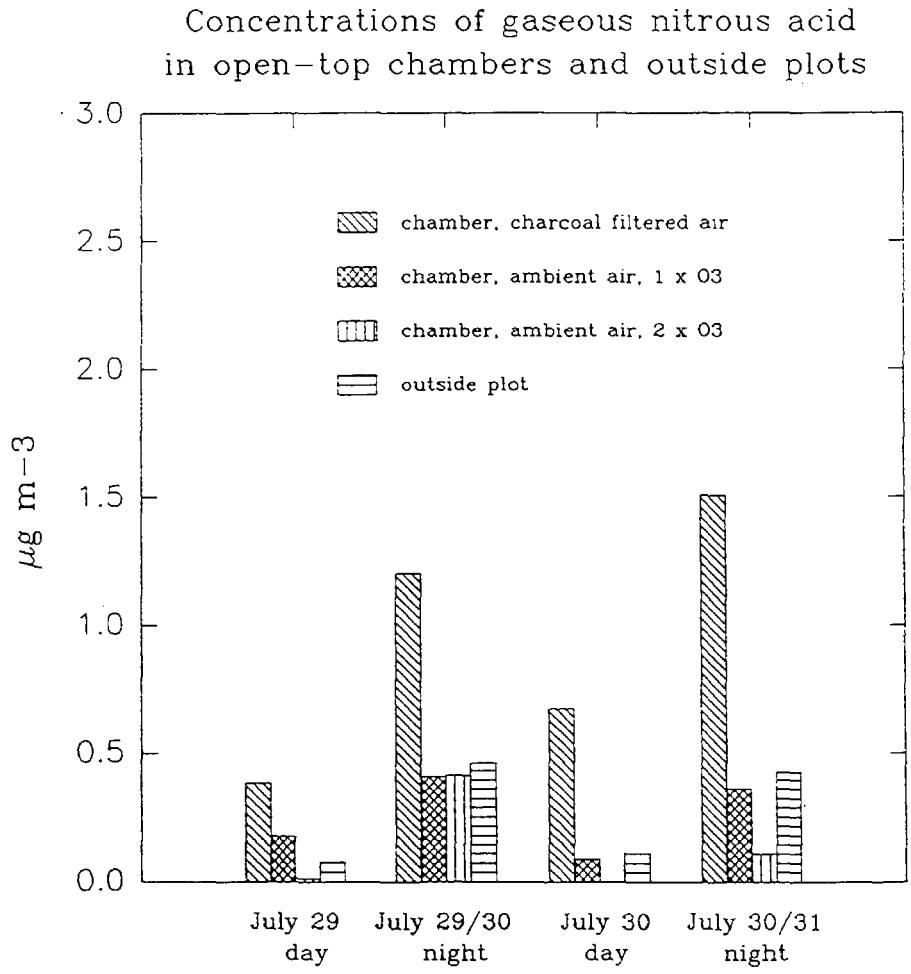

Concentrations of gaseous nitrous acid
in open-top chambers and outside plots
3.0
2.5
2.0
1.5
1.0
0.5
0.0
μg m-3
chamber, charcoal filtered air
chamber, ambient air, 1 x O3
chamber, ambient air, 2 x O3
outside plot
July 29
day
July 29/30
night
July 30
day
July 30/31
night

Table 5. Concentrations of gaseous nitrous acid in various locations (microgram/ m^3).

Location	Period and time of measurement	Range	Mean	Reference
Fortress Mt., Rocky Mtns, Alberta	3 yrs, 24 h averages	0.005-0.46	0.05	Legge and Krupa, 1989
Whitaker Forest, Sierra Nevada, CA	Summer, day " night		0.07	Temple et al., 1990
Shirley Meadow, Sierra Nevada, CA	Summer, day " night	0.00-1.94 0.06-0.64	0.36 0.19	Temple and Bytnerowicz 1991
Camp Osceola, San Bernardino Mtns, CA	Sept., 24 h		0.09	Fenn and Bytnerowicz 1992
Camp Paivika, San Bernardino Mtns, CA	Sept., 24 h		0.00	Fenn and Bytnerowicz 1992
Tanbark Flat, San Gabriel Mtns, CA	Summer, day " night	0.20-1.50 0.00-2.00	0.33 0.83	Grosjean and Bytnerowicz 1993
Goteborg, urban area, Sweden	May, 24 h		1.80	Sjodin, 1988
Claremont, Los Angeles Basin, CA	Summer	1.00-9.00	3.40	Sickles et al., 1988
Chico, CA	July, day " night		0.09 0.45	this study

Figure 3

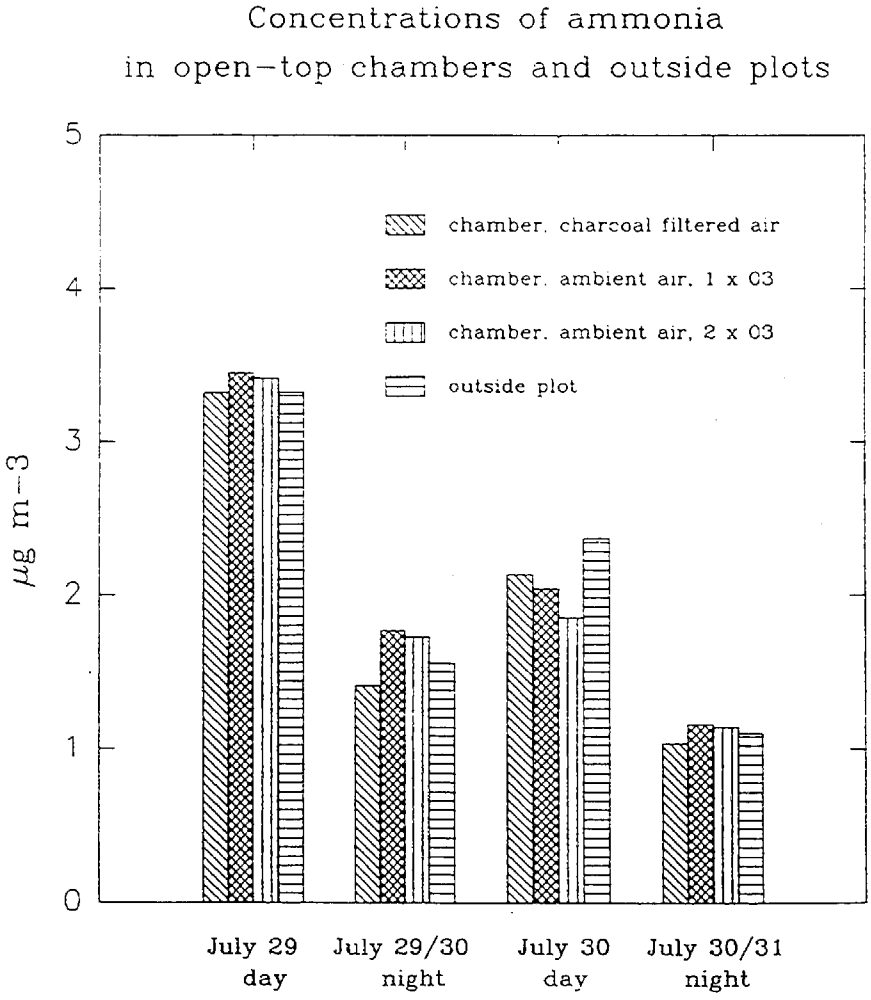
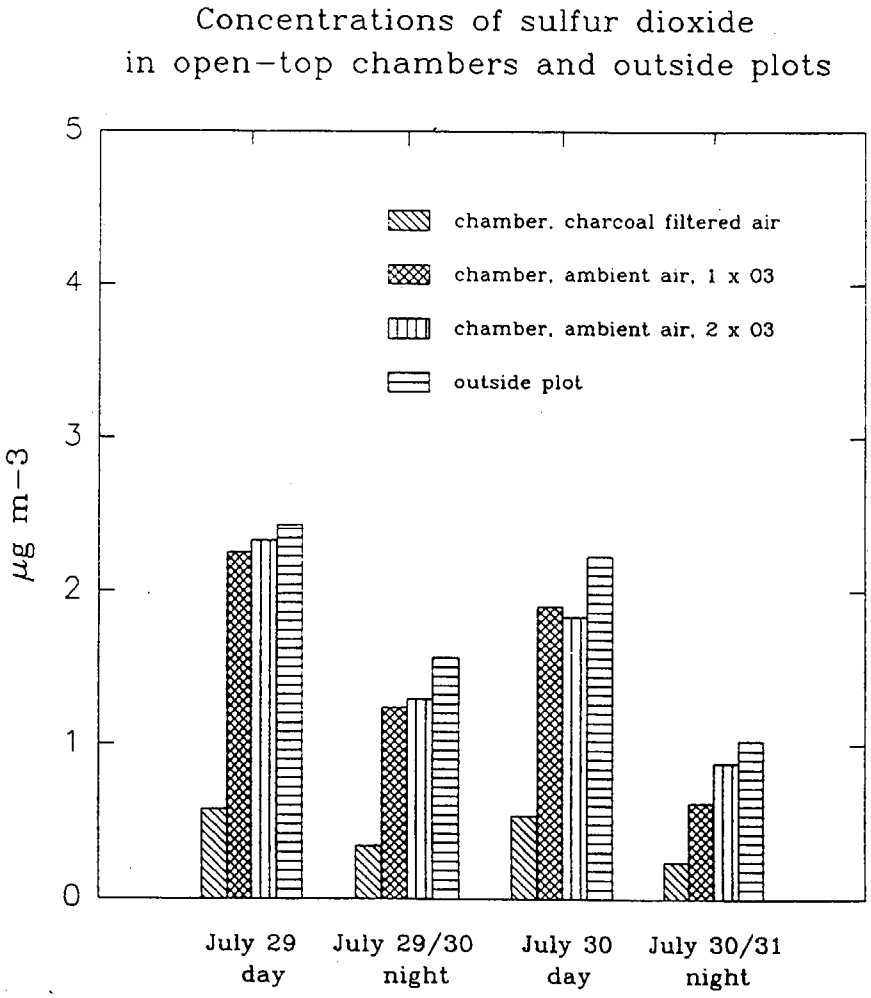

Concentrations of ammonia
in open-top chambers and outside plots
chamber, charcoal filtered air
chamber, ambient air, 1 x O3
chamber, ambient air, 2 x O3
outside plot
μg m-3
0
1
2
3
4
5
July 29
day
July 29/30
night
July 30
day
July 30/31
night

Figure 4


Concentrations of sulfur dioxide
in open-top chambers and outside plots
chamber, charcoal filtered air
chamber, ambient air, 1 x O3
chamber, ambient air, 2 x O3
outside plot
μg m-3
0
1
2
3
4
5
July 29
day
July 29/30
night
July 30
day
July 30/31
night

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