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Ozone, nitric acid, and ammonia air pollution is unhealthy for people and ecosystems in southern Sierra Nevada, California

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Ozone concentrations remained unchanged while those of nitric acid vapor and ammonia decreased along the river drainage crossing the Sierra Nevada Mountains.

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ABSTRACT

Two-week average concentrations of ozone (O_3), nitric acid vapor (HNO_3) and ammonia (NH_3) were measured with passive samplers during the 2002 summer season across the central Sierra Nevada Mountains, California, along the San Joaquin River drainage. Elevated concentrations of the pollutants were determined with seasonal means for individual sites ranging between 62 and 88 ppb for O_3, 1.0–3.8 μg m^{-3} for HNO_3, and 2.6–5.2 μg m^{-3} for NH_3. Calculated O_3 exposure indices were very high, reaching SUM00-191 ppm h, SUM60-151 ppm h, and W126-124 ppm h. Calculated nitrogen (N) dry deposition ranged from 1.4 to 15 kg N ha^{-1} for maximum values, and 0.4–8 kg N ha^{-1} for minimum values; potentially exceeding Critical Loads (CL) for nutritional N. The U.S., California, and European 8 h O_3 human health standards were exceeded during 104, 108, and 114 days respectively, indicating high risk to humans from ambient O_3.

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1. Introduction

The Sierra Nevada is one of the most elevated mountain ranges in the United States. The western slope of the Sierra Nevada rises gradually from the valley floor and has multiple deep west facing river drainages. The lower elevation western slopes consist of grasslands and foothill woodlands. Further east, as elevation increases, the landscape changes from mixed conifer forests, to alpine meadows and lakes.

The Sierra Nevada Mountains rise to the east of the San Joaquin Valley, one of the most polluted areas in the United States (American Lung Association, 2002a, 2002b, 2003). The San Joaquin Valley has a northwest to southeast orientation approximately 100 miles wide by 300 miles long. Major urban centers and agricultural areas are located to the west of the Southern Sierra Nevada. The study area is located in the Southern Sierra Nevada, on the Sierra National Forest, just south of Yosemite National Park. Fresno County, adjacent to and west of the study sites, is an agricultural center. The nearest major urban area is the city of Fresno. Air pollution is typically generated in the urban and agricultural areas of the central valley and moved toward the Sierra Nevada with a prevailing west to east wind pattern.

Various anthropogenic activities (motor vehicle traffic, energy production, agriculture, industry) and long-range transport of polluted air masses result in elevated concentrations of primary and secondary air pollutants downwind of pollution source areas (Derwent and Jenkin, 1991; McKendry, 1993). Polluted air masses from the San Joaquin Valley in California, the Sacramento and San Francisco Bay areas, and daytime upslope air flow result in high concentrations of O_3, HNO_3 and NH_3 on the western slopes of the Sierra Nevada (Van Ooy and Caroll, 1995; Bytnerowicz et al., 2002b).

Air pollution has negative effects on forests (Skarby and Sellden, 1984; Ashmore et al., 1985; Colbeck, 1985; Krupa and Manning, 1988; Caroll et al., 2003), water quality (Fenn et al., 2003), and human health (WHO, 1987; Lippmann, 1989; National Research Council, 1991). Since the 1970s, high O_3 concentrations have been

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recognized as a major phytotoxic threat to Sierra Nevada forests, especially to sensitive pines such as ponderosa (*Pinus ponderosa*) or Jeffrey (*Pinus jeffreyi*) (Miller and Millecan, 1971; Pronos et al., 1978; McBride and Miller, 1999; Alonso et al., 2002). Elevated concentrations of HNO_3 and NH_3 were also measured in the western Sierra Nevada (Bytnerowicz and Riechers, 1995) including Sequoia & Kings Canyon National Parks (Bytnerowicz et al., 2002b). Nitric acid at elevated levels may change epicuticular waxes and cause lesions on leaf cuticles; predisposing plants to the effects of droughts or pathogen attacks (Bytnerowicz et al., 1999), while high concentrations of NH_3 can also be phytotoxic (Bytnerowicz et al., 1998; Krupa et al., 2003). In addition, HNO_3 and NH_3 transported from the San Joaquin Valley into the Sierra Nevada are important sources of N deposition to the forests with negative effects on ecosystems such as alteration of plant species composition, soil acidification; elevated concentrations of nitrate (NO_3^-) in soils, streams, and ground water; and increased the susceptibility of forests to drought and fires (Bytnerowicz and Fenn, 1996; Fenn et al., 2003).

There is only limited information on spatial and temporal distribution of O_3, HNO_3, and NH_3 in the Sierra Nevada, especially in their interior and eastern parts (Fraczek et al., 2001; Bytnerowicz et al., 2002b). Consequently, the objective of this study was to characterize distribution of these pollutants across the central Sierra Nevada. Passive samplers that offer an inexpensive and accurate method for measuring concentrations of different gaseous pollutants in remote areas (Krupa and Legge, 2000) were utilized for determinations of O_3, HNO_3, and NH_3 along the San Joaquin River drainage during the 2002 summer season. It was postulated that polluted air masses from the California Central Valley could deeply penetrate into the Sierra Nevada range through the San Joaquin River drainage, and might be an important contributor to elevated O_3 concentrations and atmospheric N deposition throughout the Sierra Nevada range, including its eastern parts.

2. Material and methods

2.1. Monitoring network

An air quality monitoring network was established in the summer of 2002 to measure O_3, HNO_3, and NH_3 concentrations with passive samplers at 12 sites along the San Joaquin River drainage passing across the complex topography of the central Sierra Nevada (Fig. 1; Table 1). The monitoring sites were located on a western aspect, at least 100 m from all local roads and 200 m from main roads. Free air movement from all directions was assured, and sites exposed to continuously strong winds were avoided. Sampler stands were placed at a distance of at least 2 times the height of the tallest tree from forest edges. Sparsely dispersed smaller trees or shrubs that did not obstruct the samplers, were allowed. Passive samplers under protective caps were hung on a wooden stand about 2 m from the ground level.

Passive samplers were exposed for eight two-week long periods during the summer of 2002 from June 18, 2002 to October 9, 2002 with NH_3 measurements ending on September 25, 2002 (Table 3). During the study, the McNally fire occurred in the Sequoia National Forest during the period of July 21 to August 26, 2002 (Fig. 1). The fire burned more than 150,000 acres and affected air quality in a large area of the southern Sierra Nevada and its vicinity.

2.2. Ozone passive samplers

For monitoring O_3 concentrations, Ogawa passive samplers (Pompano Beach, Florida) were used (Koutrakis et al., 1993b). In the Ogawa samplers, nitrite (NO_2^-) on cellulose filters is oxidized by ambient ozone to nitrate (NO_3^-). The exposed filters were analyzed at the USDA Forest Service PSW Research Station in Riverside California. NO_3^- on the Ogawa sampler filters was extracted with 5 ml ultrapure H_2O added to filter storage vials. The vials were shaken for 15 min on a wrist-action laboratory shaker. To 1 mL of the aliquot, 4 mL of ultrapure water were added (5 fold dilution) and in the resulting 5 mL of solution NO_3^- concentrations were determined by ion chromatography using a Dionex 4000i. A rate of NO_3^- formation (amount of NO_3^- formed on a filter over time of exposure) served as a measure of O_3 concentration. Rates of NO_3^- formation on passive sampler monitors were compared with real-time O_3 concentration measurements with the UV absorption Thermo Environmental Model 49. The empirically derived coefficients were used for calculating O_3 concentrations from all the passive sampler sites.

2.3. Nitric acid passive samplers

Passive samplers developed by the USDA Forest Service (Bytnerowicz et al., 2002a) were used to monitor HNO_3 concentrations in this study. Ambient air is absorbed on Nylasorb nylon filter as nitrate (NO_3^-). Nylon filters containing NO_3^- from the absorbed HNO_3 were placed in 250 mL Erlenmeyer flasks into which 20 mL of ultrapure H_2O were added. The flasks were covered with Parafilm, and shaken for 15 min on a wrist-action laboratory shaker. Concentration of NO_3^- was determined with ion chromatography on a Dionex Model 4000i. Three replicate nylon filters were used at each monitoring site, and the concentrations of HNO_3 were calculated using calibration curves developed by collocating passive samplers with annular denuder systems (Koutrakis et al., 1993a).

2.4. Ammonia passive samplers

Ammonia concentration was determined with Ogawa passive samplers (Roadman et al., 2003). Ammonia was absorbed on two replicate cellulose pads coated with citric acid forming ammonium citrate. Filters were placed in vials containing 8 ml of ultrapure H_2O and shaken for 15 min on a wrist-action laboratory shaker. The ammonium concentration in filter extracts was determined colorimetrically on a Technicon Autoanalyzer and ambient NH_3 concentrations were calculated using calibration curves developed by collocating passive samplers with annular denuder systems (Koutrakis et al., 1993a).

2.5. Meteorological data

Meteorological data (June 18–October 9, 2002) was obtained from two remote automatic weather stations (RAWS) stored by the Western Regional Climate Center, and downloaded from their website (www.wrcc.dri.edu). Both weather stations are located on the San Joaquin River Drainage (Fig. 1) and are operated by the United States Forest Service (USFS). The Hurley RAWS site is located closest to the San Joaquin valley at an elevation of 369 m, while Devils Postpile RAWS (Table 1) is located on the west side of Mammoth Mountain Pass near the Starkweather monitoring station at an elevation of 2301 m.

2.6. Ozone exposure indices and N deposition

Hourly O_3 concentrations were produced for Auberry (site 1), Redinger Lake (site 2), Italian Bar (site 3), Mammoth Pool Powerhouse (site 4), Fish Creek (site 10), and Starkweather Lake (site 11) using the statistical approach of coupling passive O_3 data with meteorological variables described by Krupa et al. (2003). Hurley meteorological data was used for sites 1, 2, 3, and 4 because the Hurley site was most representative of the meteorology at these sites. Meteorology data from Devils Postpile was used as the most representative for sites 10 and 11. Hourly data was then used to calculate the different ozone exposure indices below. Hourly O_3 concentrations were not produced for the other sites due to a lack of representative meteorological data.

Two predictive models were considered: (1) a nonlinear-polynomial model with the first and second degree powers for the passive O_3 sampling, and (2) a linear model. The models used the following variables: temperature (T), relative humidity (RH), hour of the day (Hour), wind speed (WS), and O_3 passive concentration (O_3p). Real-time hourly O_3 concentrations for the Shaver Lake site were obtained from the California Air Resources Board. Ozone concentrations at that site were measured with the UV absorption instrument Dasibi Model 1003AH Ozone Analyzer. Passive monitoring equipment was collocated with this real-time equipment for the entire study. The two-week O_3 measured with the passive equipment highly correlated ($R^2 = 0.814$) with the two-week average calculated using the hourly data at Shaver Lake over the eight-week sampling period. The Shaver Lake site was used for model calibration only. The linear model ($r = 0.795$, significance <0.001) was selected because it performed better than the nonlinear model ($r = 0.644$, significance <0.001) when compared with the real-time hourly O_3 concentrations. For the study period, hourly O_3 at Shaver Lake had a mean of 56 ppb, a median of 53 ppb, a standard deviation of 20 ppb, a minimum of 8 ppb, and a maximum of 130 ppb. Linear model calculation for this time using the Shaver Lake passive site had a mean of 55 ppb, a median of 54 ppb, a standard deviation 17 ppb, a minimum of 19 ppb, and a maximum of 89 ppb. The stepwise regression used to build the predictive model excluded wind speed because it was not significant. The prediction equation is as follows: O_3 continuous $= 21.748 + 0.640\,T - 0.234\,RH + 0.370\,Hour + 0.057\,O_3p$. Model summary: $r = 0.795$, $R^2 = 0.644$, Adj. $R^2 = 0.643$, S.E. of the estimate $= 12.492$, significance <0.001. The model used in this paper had very similar predictive power as the one described by Krupa et al. (2003). Overall, the model performed well, with a very high correlation coefficient. The model did under predict the highest hourly ozone values at Shaver Lake.

Hourly values from the linear model were calculated into two-week averages and compared to the two-week values measured by the passive samplers for sites 1–4 and 10–11. In this comparison, the model did not perform as well ($R^2 = 0.477$). The authors feel comfortable using the linear model as a conservative estimate of ozone exposure because the model under-predicted the two-week average ozone data (Table 6) at all locations during almost all collection periods. It is likely that the

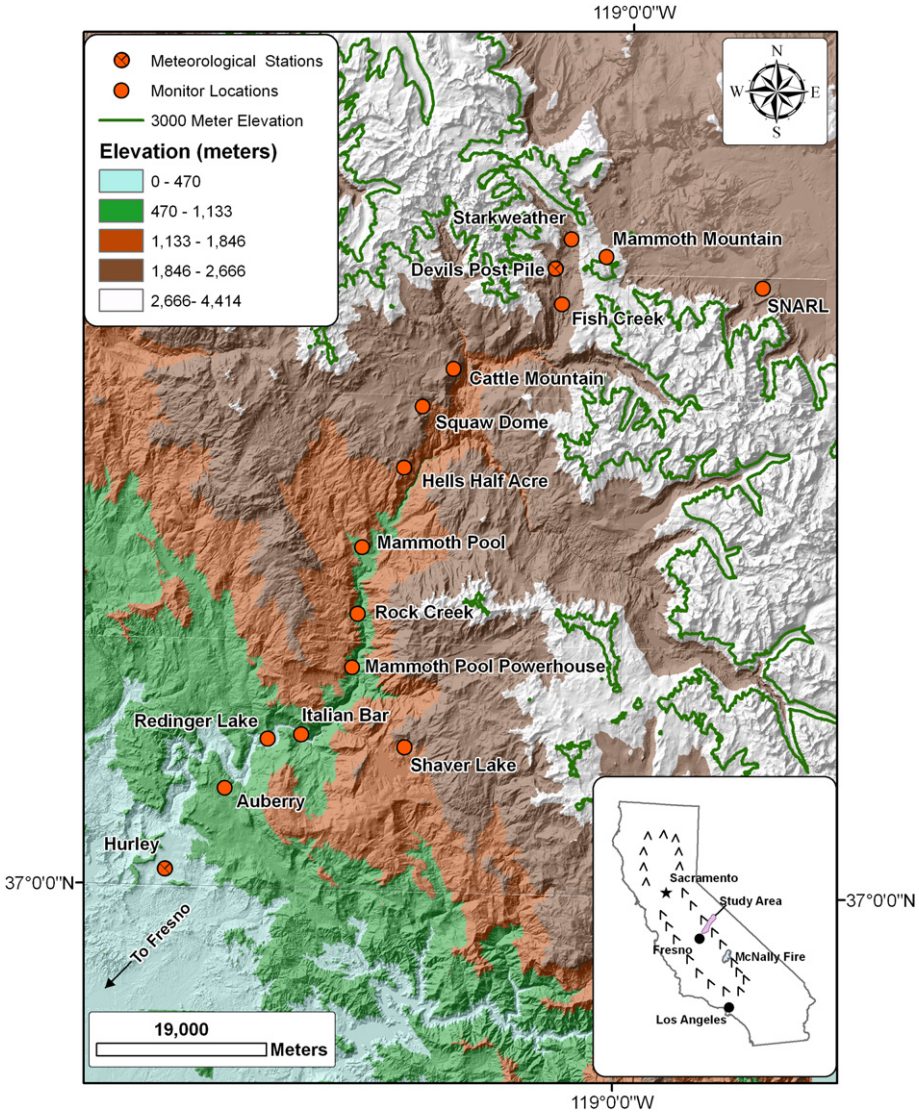

Fig. 1. Location of the monitoring stations along the San Joaquin River drainage in the central Sierra Nevada, California. Please see Table 1 for more information about the monitoring sites.

Table 1
Coordinates and Altitude of the Sampling Locations.

Site Name (Site Number)	Latitude (DD)	Longitude (DD)	Altitude (M)
Auberry (1)	37.099	−119.496	680
Redinger Lake (2)	37.150	−119.442	542
Italian Bar (3)	37.155	−119.400	513
Mammoth Pool Powerhouse (4)	37.224	−119.338	882
Rock Creek (5)	37.278	−119.333	868
Mammoth Pool (6)	37.345	−119.329	1106
Hells Half Acre (7)	37.426	−119.279	1908
Squaw Dome (8)	37.488	−119.257	2078
Cattle Mountain (9)	37.527	−119.219	2275
Fish Creek (10)	37.594	−119.084	2241
Starkweather Lake (11)	37.660	−119.073	2446
Mammoth Mountain (12)	37.643	−119.029	2933
SNARL	37.614	−118.830	1250
Shaver Lake	37.138	−119.267	1700
Hurley (RAWS)	37.015	−119.567	369
Devils Postpile (RAWS)	37.63	−119.093	2301

DD = Decimal Degrees.
M = Meters.
RAWS = Remote Automatic Weather Stations.

predicted ozone exposure indices are higher than what it is presented here. The largest differences between the observed and the predicted two-week averages happened during the months of July and August (Table 6). Although the models presented here are not perfect, they are helpful in predicting exposure regimes in mountain locations where topography, lack of power, and access restrict the availability of real-time data.

Ozone exposure indices (SUM00, SUM60, SUM70, SUM80, W126, and SOMO35) and 8-hour (h) human health standards (U.S. (75 ppb), California (70 ppb), and Europe (50 ppb)) were calculated for approximately four months (June 18–October 9, 2002). A new secondary standard proposed by the U.S Environmental Protection Agency, W126 for 12 h between 08:00-20:00 PST for 3 months (July 1–September 30), was also calculated. The SOMO35 metric and the 8 h standards are used to protect human health while the others are used to protect vegetation. The SUM00 is the sum of all hourly concentrations without a threshold. SUM60, SUM70, and SUM80 are the sums of all hourly concentrations above 60 ppb, 70 ppb, and 80 ppb respectively. The W126 is a sigmoidally weighted index (Lefohn and Runeckless, 1987) where higher concentrations have a greater weighing. The SUM60 and W126 have been suggested as the most acceptable and most commonly used vegetation exposure indices in the U.S. (U.S. EPA, 1996; Musselman et al., 2006). The SOMO35 is the sum of excess 8 h mean above 35 ppb (Amann et al., 2005).

Estimates of N dry deposition from HNO_3 and NH_3 were done by the inferential method utilizing two-week-averaged concentrations of the pollutants and the literature deposition velocity values (V_d): minimal ($HNO_3 = 2$ cm s^{-1}, $NH_3 = 1.8$ cm s^{-1}) and maximal ($HNO_3 = 7$ cm s^{-1}, $NH_3 = 2.6$ cm s^{-1}) (Hanson and Linberg, 1991). The deposition values taken from the literature account for vegetation, wind velocities,

and exposure, but there are still numerous uncertainties related to these deposition velocities and the expected N deposition. The authors decided to use the minimal and the maximal deposition velocities values to provide a range of dry N deposition. Therefore, the range of values presented here are a first step into understanding potential scenarios of N deposition to the forests. Uncertainties from such inferential methods are apparent and will likely be reduced in the future as N deposition is more thoroughly studied. The method use here is a starting point in the understanding of exposure and risk to the forests. The estimated N deposition values for the duration of the study were used for evaluation of potential exceedances of Critical Loads (CL) for nutritional N. Ammonia data was not collected at sites 10 and 11, and HNO_3 and NH_3 data was not collected at site 12.

2.7. Statistical tests

Statistical analyses were conducted using SPSS 11.5 (SPSS™). Differences in O_3, HNO_3, and NH_3 concentrations between sites and periods of collection were determined using analysis of variance (one way ANOVA). The Tukey test (multiple comparison procedure) *post hoc* analysis was used for comparison between sites and between collection periods. A confidence interval of 95% ($\alpha = 0.05$ level of significance) was used for analysis of significant difference.

3. Results

3.1. Ozone

Mean O_3 concentrations for individual monitoring sites in the San Joaquin drainage were not significantly different (Table 2), while two-week O_3 values ranged from 41 ppb in period 4 at Starkweather Lake to 186 ppb in period 5 at Squaw Dome (Fig. 2, Table 2). A significant effect of time ($P < 0.001$) was found with the highest concentrations recorded during August 14–28 (Fig. 2). That period (Period 5) was significantly different from other periods ($P < 0.05$) (Table 3) and coincided with the latter period of the McNalley fire, which started on July 21st and ended on August 26th 2002. During that period, extremely high concentrations of O_3 were recorded in some locations along the San Joaquin River drainage including 186 ppb in Squaw Dome and 132 ppb on Mammoth Mountain (Fig. 2); at the time other southern Sierra Nevada locations showed significantly elevated O_3 levels (Cisneros et al., 2007). These very high concentrations were caused by increased production of O_3 precursors (NOx, CO, and VOCs) emitted from the McNally fire (Cheng et al., 1998; Sandberg et al., 1999; Cisneros et al., 2007, unpublished). During periods 6–8 (August 28–October 9) there was a tendency toward lower O_3 concentrations, similar to another O_3 monitoring campaign in the Sierra Nevada (Fraczek et al., 2001; Bytnerowicz et al., 2002b), due to less intensive photochemical reactions at lower temperatures and solar radiation.

3.2. Nitric acid

The highest HNO_3 concentrations were recorded at Auberry (seasonal mean = 3.8 μg m^{-3}) and the lowest at Cattle Mountain (seasonal mean = 1.0 μg m^{-3}), while the two-week concentrations for all sites ranged from 0.5 to 4.7 μg m^{-3} (Table 2). Sites in the San Joaquin River drainage were found to be significantly different ($P < 0.001$) with the highest HNO_3 concentrations near the San Joaquin Valley that gradually decreased eastwards (Table 2, Fig. 2). A significant effect of time ($P = 0.026$) on HNO_3 concentration occurred (Table 3). Periods 4 and 5 were characterized by the highest mean HNO_3 concentrations (2.5 and 2.4 μg m^{-3}, respectively), which could be related to the McNally fire and increased NO_x emissions (Cheng et al., 1998) promoting HNO_3 formation (Urbanski et al., 2009). The lowest values of the season (seasonal mean = 1.1 μg m^{-3}) were recorded in the fall (Period 8), similar to what was observed in the Sequoia National Park study in 1999 (Bytnerowicz et al., 2002b).

3.3. Ammonia

Ammonia concentrations on the San Joaquin River transect were the highest at Auberry (seasonal mean = 5.2 μg m^{-3}), and lowest at Cattle Mountain (seasonal mean = 2.6 μg m^{-3}). Concentrations between the sites were significantly different ($P < 0.001$), and gradually and significantly decreased with distance from the San Joaquin Valley (Table 3, Figs. 2 and 5). Significant effect of time ($P < 0.001$) on NH_3 concentrations was also determined with the highest value (5.4 μg m^{-3}) recorded in period 6, and the lowest (2.7 μg m^{-3}) during period 1 (Table 3, Fig. 2).

3.4. Meteorological observations

Meteorological observations are shown for two sites, Hurley and Devils Postpile (Fig. 1). Both sites have well defined diurnal wind patterns that are typical of mountain valley and canyon locations (Stull, 1988; Van Ooy and Caroll, 1995). Fig. 3 shows the mean hourly values of wind speed, direction, temperature, and relative humidity for the duration of the study. At both sites, winds were southwesterly during the day, changing to south–southeasterly at night, reflecting the influence of local topography.

Wind speed also showed a distinct diurnal variation. Wind speed for both sites increased at about 0800 PST and decreased around 1900 PST. The nighttime winds were near calm with mean speed less than 0.5 m s^{-1}, while, during the daytime, wind speeds

Table 2
Two-Week Average O_3, HNO_3, and NH_3 Concentrations in the San Joaquin River Drainage during the summer of 2002.

Site Name	O_3 (ppb)		HNO_3 (μg/m^3)		NH_3 (μg/m^3)	
	Range	Mean (S.D.)	Range	Mean (S.D.)	Range	Mean (S.D.)
Auberry	65–89	77(10)a	2.2–4.7	3.8(1.0)a	4.3–7.3	5.2(1.0)a
RedingerLake	62–98	83(12)a	2.3–4.2	3.2(0.6)ab	3.3–6.3	4.7(1.0)ab
Italian Bar	56–95	77(12)a	1.2–4.0	2.4(0.9)bc	2.9–6.8	4.2(1.3)abc
Mammoth Pool Powerhouse	62–97	81(11)a	1.5–3.3	2.4(0.6)bc	2.0–6.9	3.7(1.7)abc
Rock Creek	56–92	70(11)a	0.8–2.8	1.7(0.6)cd	2.0–4.7	3.0(0.9)bc
Mammoth Pool	49–82	69(11)a	0.6–2.7	1.5(0.6)cd	1.9–4.7	3.2(1.0)bc
Hells Half Acre	63–95	77(10)a	0.6–1.6	1.3(0.3)d	2.6–4.4	3.3(0.7)bc
Squaw Dome	60–186	88(41)a	0.5–1.4	1.1(0.3)d	2.2–4.3	3.0(0.7)bc
CattleMountain	60–94	75(13)a	0.6–1.4	1.0(0.3)d	1.6–3.7	2.6(0.9)c
Fish Creek	58–94	71(14)a	0.7–1.8	1.5(0.4)cd		
StarkweatherLake	41–88	62(13)a	0.6–3.2	1.5(1.0)cd		
MammothMountain	58–132	82(23)a				
SNARL	46–76	59(11)				
Shaver Lake	47–123	73(24)				
P value	0.280		<0.001		<0.001	

Different letters following mean and S.D. indicate significant differences ($P < 0.05$) between sites as determined with the Tukey test. SNARL and Shaver Lake were not included in determining differences between sites.

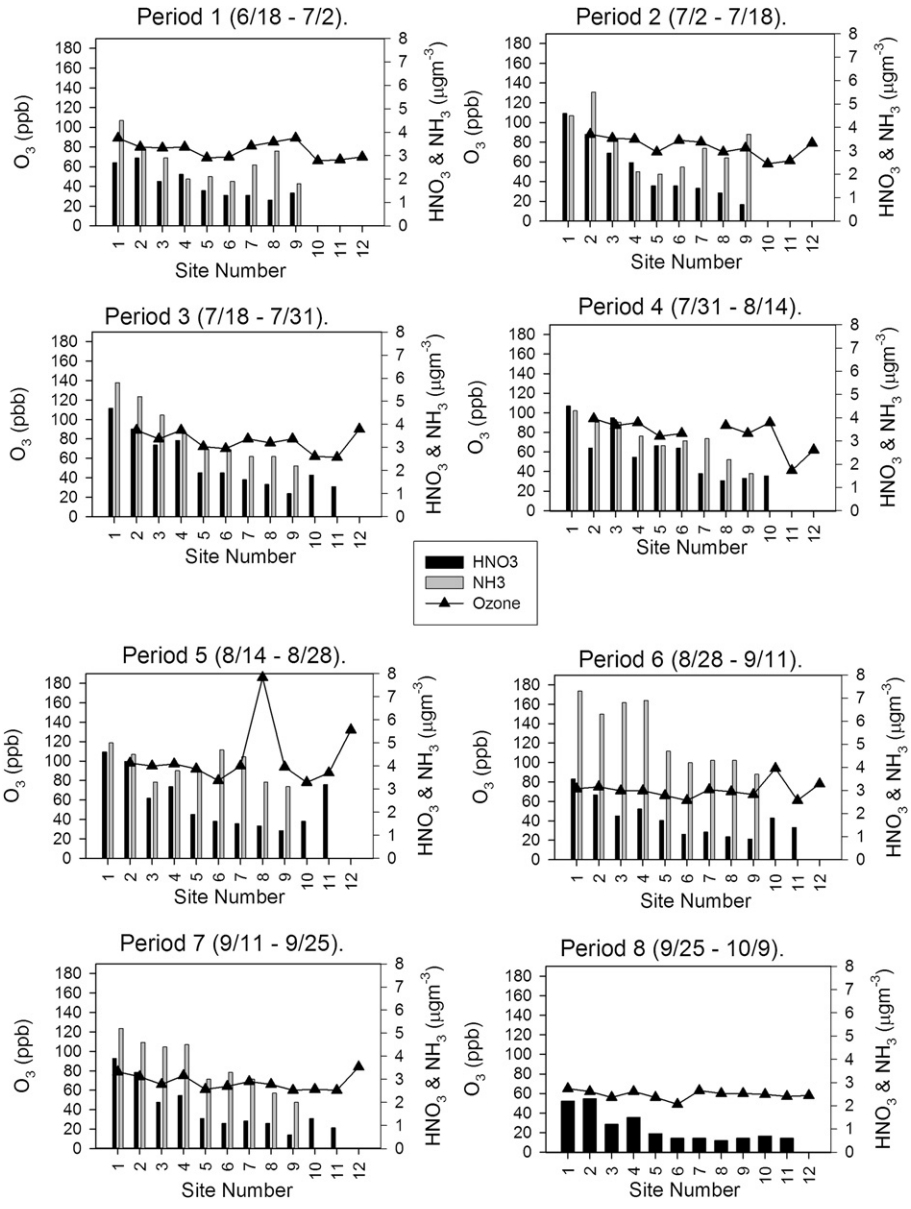

Fig. 2. Temporal distribution of two-week long average O_3, HNO_3, and NH_3 concentrations in the San Joaquin River drainage during the 2002 summer.

Table 3
Passive samplers (O_3, HNO_3, and NH_3) two-week periods, Summer 2002.

Collection Period (Date)	Results of Tukey's test		
	O_3 (ppb)	HNO_3 (µg/m^3)	NH_3 (µg/m^3)
	Results (Mean)	Results (Mean)	Results (Mean)
1 (6/18–7/2)	(77)b	(1.8)ab	(2.7)b
2 (7/2–7/18)	(75)bc	(2.2)ab	(3.3)b
3 (7/18–7/31)	(77)bc	(2.3)ab	(3.6)b
4 (7/31–8/14)	(79)b	(2.5)a	(3.1)b
5 (8/14–8/28)	(103)a	(2.4)a	(4.0)ab
6 (8/28–9/11)	(72)bc	(1.8)ab	(5.4)a
7 (9/11–9/25)	(68)bc	(1.7)ab	(3.6)b
8 (9/25–10/9)	(59)c	(1.1)b	
P value	<0.001	0.026	<0.001

Note: NH_3 collection ended 9/25.
Different letters following mean and S.D. indicate significant differences ($P < 0.05$) between periods as determined with the Tukey test.

were generally 1–2 m s^{-1} as the result of downward mixing of higher winds aloft by the daytime growth of the convective boundary layer. The mean wind speed at Hurley was higher than wind speed at Devils Postpile at all hours over a diurnal cycle. This was unexpected because wind speed in the lower atmosphere generally increases with elevation. However, the weaker winds at Devils Postpile may be due to blocking affects of local topography compared to the more open site location at Hurley. Stronger winds at Hurley were more variable than those at Devils Postpile as indicated by higher standard deviation values (Table 4).

3.5. Ozone exposure indices

Calculations of the O_3 exposure indices were performed for six sites (sites 1–4, and 10–11) because only these sites had the meteorological data required to compute the metrics.

The sum of all hourly average concentrations above 0, 60, 70, and 80 ppb were in the range of 140–191, 80–151, 54–125, and 12–187 ppm h for SUM00, SUM60, SUM70, and SUM80, respectively.

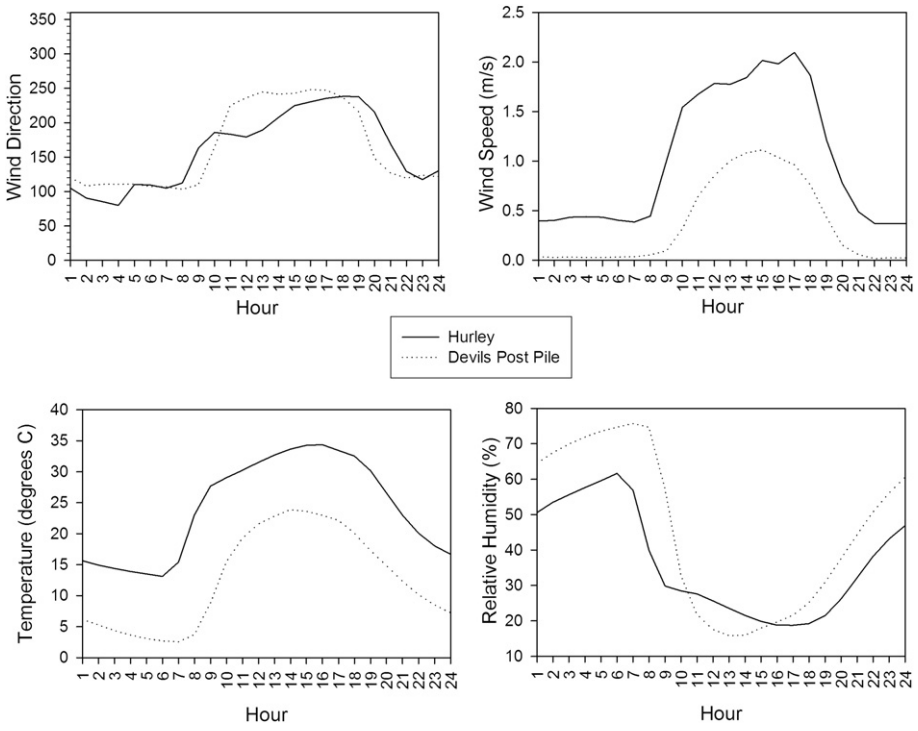

Fig. 3. Hourly averaged meteorological data from June, 18 through October, 9 2002.

The values of all of the SUMxx metrics decreased with distance from the Central Valley, with sites 2–4 experiencing the highest exposures (Fig. 4). Site 1 experienced slightly lower O_3 exposures indices than sites 2–4 which can be explained by the fact that Auberry is near the San Joaquin Valley and does not experience the same canyon influences as sites 2–4. In general, sites 1–4 experience more than twice the values of sites 10 and 11. The W126 indices were in the range of 57–124 ppm h. The W126 indices in sites 1–4 were about 2 fold higher than the calculated indices for sites 10 and 11. The W126 index for site 1 was similar to those described above for the SUMxx when compared to sites 2–4 (Fig. 4).

All sites were above the proposed U.S. secondary standard *W126 threshold of 7–15 ppm h. The *W126 indices were in the range of 46–85 ppm h. Sites 1–4 calculated indices were higher than sites 10 and 11.

The number of days when the daily maximum 8 h average concentrations exceeded the U.S. (75 ppb), California (70 ppb), and European (50 ppb) standards were in the range of 42–104, 81–108, and 110–114 respectively for the selected sites (Table 5). Sites 1–4 experienced the greatest count of exceedances of the U.S. and California 8 h standard, while for the European 8 h standard all sites had a similar count of exceedances.

The sum of excess of daily maximum 8 h average concentrations greater than 35 ppb (SOMO35) was in the range of 4161–5748 ppb day for all sites (Table 5). Similar to the W126 index, sites 1–4 experienced higher SOMO35 values than sites 10 and 11.

3.6. N deposition from nitric acid and ammonia

Calculated maximum and minimum dry N deposition from HNO_3 and NH_3 decreased eastwards (Table 7). Sites closest to the San Joaquin Valley had the highest levels of HNO_3 and NH_3 deposition, with much lower N deposition resulting from HNO_3 than from NH_3. Deposition of N from NH_3 for all sites accounted for 62–76% and 79–88% of the calculated maximum and minimum N depositions respectively. The calculated maximum N deposition values from HNO_3 in all sites ranged between 1.4 kg N ha^{-1} and 5.8 kg N ha^{-1}, whereas the calculated minimum values were

Table 4
Descriptive statistics for hourly meteorological data in the San Joaquin River Drainage from June 18th through October 9th, 2002.

		T	RH	WD	WS	U	V
Hurley	Mean	24	36	325	1.0	0.008	−0.012
	S.D.	9	18	224	0.8	0.903	0.948
	Range	3–43	10–100	–	0.0–4.5	–	–
Devils Postpile	Mean	13	46	196	0.4	−0.006	−0.023
	S.D.	9	27	226	0.5	0.443	0.433
	Range	−3 to 35	7–100	–	0.0–2.2	–	–

T = Temperature (degrees C).
RH = Relative Humidity (%).
WD = Wind Direction (Degrees).
WS = Wind Speed (m/s).
U = east-west wind component (m/s).
V = north-south wind component (m/s).

Table 5
Count of exceedences of the U.S. Federal 8 h O_3 standard (75 ppb), California 8 h O_3 standard (70 ppb), European 8 h O_3 standard (50 ppb). Table also presents the results of calculated SOMO35 (ppb day).

Site	8 h mean ≥ 75 ppb	8 h mean ≥ 70 ppb	8 h mean ≥ 50 ppb	SOMO35
Auberry	104	108	114	5459.732
Redinger Lake	104	108	114	5747.981
Italian Bar	103	108	114	5713.85
Mammoth Pool Powerhouse	104	108	114	5736.644
Fish Creek	43	85	110	4217.729
Starkweather Lake	42	81	110	4160.729

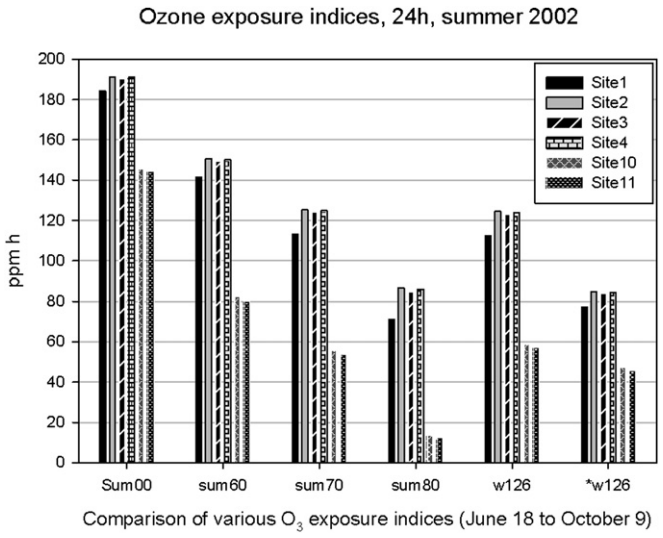

Fig. 4. Ozone exposure indices for mountain locations (Sites 1–4 and 10–11) in summer 2002 (June, 18–October, 9). Note: w 126 is calculated for a 24 h period. *w126 refers to the proposed EPA standard calculated for a 12 h period (8am–8pm).

between 0.4 kg N ha^{-1} and 1.7 kg N ha^{-1}. Nitrogen deposition from NH_3 in the calculated maximum values ranged from 4.7 to 9.5 kg N ha^{-1}, whereas the calculated minimum values ranged from 3.2 to 6.6 kg N ha^{-1}. The range of total N deposition in the calculated maximum values for all sites was 1.4–15 kg N ha^{-1}, while the calculated minimum values were 0.4–8 kg N ha^{-1}.

4. Discussion

4.1. Pollutants across the Sierra Nevada

Very high O_3 concentrations in southwestern Sierra Nevada locations are caused by polluted air masses from the California Central Valley (Van Ooy and Caroll, 1995). Along the San Joaquin River drainage, locations closer to the valley experienced only slightly higher levels of O_3 than locations up canyon (Fig. 2). The differences between the individual sites were not statistically significant (Table 2). Ozone concentrations did not significantly change with increased elevation (Fig. 5). Similarity in elevated O_3 concentrations along the San Joaquin River drainage transect is an indication of long-range transport of air pollutants from California's Central Valley to the east side of the Sierra Nevada. Pollutants along the drainage (~2400 m) are transported through Mammoth Pass (2790 m, located south of Mammoth Mountain) and through the Minarets Vista Point area (2800 m, located north of Mammoth Mountain). The relatively low elevation of these two locations along the Sierra Nevada crest allows the polluted San Joaquin Valley air masses rising up the canyon to funnel across to the east side of the Sierra Nevada (Fig. 1). Topography in these locations allows for up canyon or up valley winds to outflow from the stable core and the capping inversion formed by the drainage topography (Stull, 1988). Elevated levels of O_3 found at SNARL (Table 2), a site east of Mammoth Pass, is another indication of air pollutant transport from the San Joaquin Valley across the Sierra Nevada.

The RAWS stations (Hurley and Devils Postpile) supplied more evidence to support the hypothesis that O_3 and other pollutants are transported through the San Joaquin River drainage across the Sierra Nevada. Winds from both sites were predominantly up canyon in the east-northeast direction between 10:00 and 20:00 PST for the duration of the study (Fig. 3). The valley wind speeds recorded at Hurley RAWS station are strong enough to reach the RAWS station at the Devils Postpile site in less than 10 h (wind speeds > 2 m s^{-1}). The weak down canyon or down drainage winds are not strong enough to re-circulate polluted air masses to the Central Valley, which suggests that the pollutants are kept in the drainage at night and then moved by advection during the day up drainage in an east-northeasterly direction.

HNO_3 concentrations decreased significantly with distance from the San Joaquin Valley and elevation along the San Joaquin River Drainage (Fig. 5). This is due to HNO_3 high deposition velocity to surfaces and stomatal uptake by vegetation (Hanson and Linberg, 1991). Similar to HNO_3, concentration of NH_3 also decreased with increasing elevation and distance from the San Joaquin Valley (Fig. 5). Although surface deposition of NH_3 is lower than HNO_3, its stomatal uptake is higher than HNO_3 (Hanson and Linberg, 1991). The reduction of HNO_3 and NH_3 concentrations further up the San Joaquin River drainage could also be explained by the reaction between these compounds that leads to NH_4NO_3 formation (Blanchard et al., 1999). Therefore, no significant amounts of HNO_3 and NH_3 could be transported to the east side of the Sierra Nevada because of their high deposition velocity and reactivity.

4.2. Effects of fire

During period 5 (August 14–28) of this study, O_3 levels were higher than during other collection periods. This high two-week O_3 episode is attributed to the McNally fire that started on July 21st and ended on August 26th 2002 (Cisneros et al., 2007, unpublished). During that period, extremely high concentrations of O_3 were recorded in the San Joaquin River drainage (186 ppb in Squaw Dome and 132 ppb on Mammoth Mountain) (Table 2, Fig. 2) with elevated O_3 levels present at other sites in the southern Sierra Nevada (Cisneros et al., 2007,

Table 6
Passive ozone (ppb) two-week average observed values versus two-week average values predicted by the model.

Date	Auberry		Redinger		Italian Bar		Mammoth Pool		Fish Creek		Starkweather	
	O	P	O	P	O	P	O	P	O	P	O	P
(6/18–7/2)	89	70	80	70	79	70	80	70	66	53	67	53
(7/2–7/18)		71	88	76	84	76	83	76	58	57	61	57
(7/18–7/31)		69	89	74	80	73	89	74	62	57	61	57
(7/31–8/14)		67	94	73	87	72	90	73	90	58	41	55
(8/14–8/28)		66	98	72	95	72	97	72	78	58	88	59
(8/28–9/11)	73	69	75	69	71	69	71	69	94	53	61	51
(9/11–9/25)	79	70	74	69	66	69	75	70	61	53	60	53
(9/25–10/9)	65	61	62	61	56	61	62	61	59	45	57	45
Average	77	68	83	70	77	70	81	70	71	54	62	54
S.D.	10	3	12	5	12	5	11	5	14	4	13	4

O = Observed.
P = Predicted.

Table 7
Range of calculated dry N deposition (kg N ha^{-1}) from HNO_3 and NH_3 concentrations.

Site Number	HNO_3	NH_3	Total N
1	1.7–5.8	6.6–9.5	8.2–15.3
2	1.4–4.8	6.0–8.6	7.3–13.4
3	1.1–3.7	5.2–7.5	6.3–11.2
4	1.0–3.7	4.7–6.8	5.7–10.4
5	0.7–2.5	3.8–5.5	4.5–8
6	0.6–2.2	4.0–5.8	4.6–8
7	0.6–2	4.1–6	4.7–7.9
8	0.5–1.7	3.7–5.4	4.2–7.1
9	0.4–1.5	3.2–4.7	3.7–6.2
10	0.5–1.6	No data collected	0.5–1.6
11	0.4–1.4	No data collected	0.4–1.4

unpublished). Apparently this episode of elevated ambient O_3 levels were caused by increased emissions of the O_3 precursors from the McNally fire as has been reported for other forest fires (Cheng et al., 1998; Sandberg et al., 1999).

Elevated concentrations of HNO_3 were recorded at some sites in the San Joaquin River drainage (sites 3, 5, 6, and 11) during periods 4–5 (July 31–August 28 (Fig. 2)). The values recorded during periods 4–5 were significantly greater than the values recorded for other periods (Table 3, Fig. 2). These high values could be attributed to the increased generation of HNO_3 from the NO_x emissions of the McNally fire (Tarnay et al., 2001; Cisneros et al., 2007).

During periods 5–6 (August 14 to September 11), NH_3 concentrations were significantly higher than in any other period (Table 3), including the sites far away from agricultural activities of the San Joaquin Valley (Fig. 2). Forest fires are well-known to emit significant amounts of NH_3 during the combustion of fuels and in the smoldering phase (Andrae and Merlet, 2001; Dennis et al., 2002; Clinton et al., 2006; Yokelson et al., 2007). The McNally fire was declared to be out on 26 August, but fires in the Sierra Nevada can continue to smolder for several weeks until they are completely out. Therefore, the increase of NH_3 during periods 5–6 could be attributed to later stages of the McNally fire when the smoldering phase is dominant.

Concentration of soil ammonium (NH_4^+) can increase greatly after fires (Turner et al., 2004; Certini, 2005), sometimes more than of an order of magnitude. Such an increase may be caused by soil heating and by NH_4^+ being incorporated to soil from ash. Concentrations of NH_4^+ may remain elevated in soil as a result of both the increase of its production and decrease in NH_4^+ consumption by plant and microbes (Fisher and Binkley, 2000). Volatilization of elevated pools of NH_4^+ from soils could additionally contribute to elevated ambient NH_3 concentrations.

4.3. Comparisons of pollutants with other locations

Ozone concentrations at the San Joaquin River drainage were elevated and typical for locations on or near drainages or canyons in the southern Sierra Nevada (Bytnerowicz, 2005). Ozone concentrations found along the San Joaquin River transect (41–186 ppb) were generally higher than those found at high elevation sites of Sequoia National Park in the 1999 summer season (40–85 ppb) (Bytnerowicz et al., 2002b). High O_3 concentrations were present along the San Joaquin River drainage throughout the season (Table 2, Fig. 2). Although lower than on the San Joaquin River transect, concentrations of O_3 were also elevated in the eastern Sierra Nevada. Mean O_3 concentrations at some sites in the San Joaquin River drainage were similar or higher than the seasonal mean O_3 concentration of 76 ppb experienced at Barton Flats, a moderately polluted site in the eastern San Bernardino Mountains in southern California (Bytnerowicz et al., 2002b). However, other sites on the western side of the San Bernardino Mountains experienced much higher O_3 levels (Bytnerowicz et al., 2008).

Nitric acid concentrations were similar to the values determined at Ash Mountain in Sequoia National Park (Bytnerowicz et al., 2002b). In the Sequoia National Park study, HNO_3 values ranged from 0.4 to 4.2 μg m^{-3}, whereas in the San Joaquin River drainage, HNO_3 ranged from 0.6 to 4.7 μg m^{-3}. The values recorded in Sequoia National Park were similar to the values recorded at Barton Flats in the San Bernardino Mountains, which is considered a moderately polluted site (Bytnerowicz et al., 1999, 2002b). In general, the HNO_3 concentrations were elevated over the Sierra Nevada background level of 0.4–0.5 μg m^{-3} (Bytnerowicz and Fenn, 1996).

Ammonia concentrations were similar to the concentrations recorded at the Ash Mountain site in Sequoia National Park (Bytnerowicz et al., 2002b). Concentrations of NH_3 were high, reflecting proximity to the agricultural lands of the San Joaquin Valley, which emit high amounts of NH_3. Ammonia concentrations were above the Sierra Nevada background levels (~0.5 μg m^{-3}) (Bytnerowicz and Fenn, 1996). Seasonal mean NH_3 in the San Joaquin River drainage was higher than the concentrations determined in the San Bernardino Mountains (Bytnerowicz and Fenn, 1996; Bytnerowicz et al., 2002b).

4.4. Potential negative effects on forests

The different O_3 exposure indices calculated for this study, used both in the United States and in Europe, indicate that phytotoxic effects to pines and other species are expected in the San Joaquin

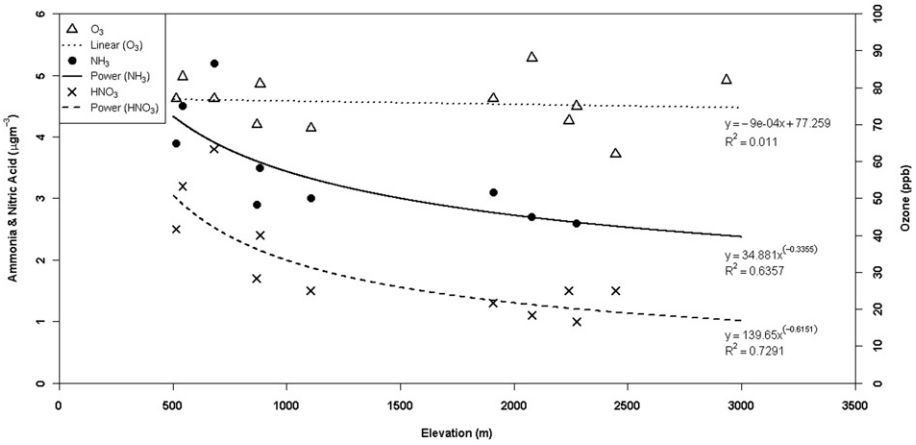

Fig. 5. Relationship between seasonal means of O_3, HNO_3 and NH_3 concentrations and elevation on the San Joaquin River transect.

River drainage and its vicinity. All of the O_3 indices indicated that sites 1–4 (closest to the California Central Valley) had much higher exposure regimes than sites 10 and 11 (near the eastern Sierra Nevada); mainly due to a greater proportion of high concentrations at sites near the Central Valley. Most of the O_3 exposure indices at sites 1–4 were similar to the very high values reported in 2002 for the Crestline (CR) site in the San Bernardino Mountains of southern California (Bytnerowicz et al., 2008). At the CR site, the O_3 exposure indices (SUM00-190 ppm h, SUM60-147 ppm h, and W126-125 ppm h) were calculated for a full 4 months (June 1–September 30), and were the highest among all studied mountainous regions in the U.S. (Bytnerowicz et al., 2008). The SUM00, SUM60, and W126 values reported in this study for sites 1–4 (~190, ~150, and ~125 ppm h, respectively) were obviously also very high, especially considering they were calculated for less than a full 4 months (June 18–October 9). These values could feasibly increase by 7–8% with a full 4 month data set. The SUM00, SUM60, and W126 indices calculated for sites 10–11 (~145, 80, and 58 ppm h respectively) were also high; showing high O_3 phytotoxic potential in the interior Sierra Nevada. All sites would be declared non-attainment under the newly proposed U.S. *W126 secondary standard.

At the beginning of the study (June 18–July 18) two-week average O_3 concentrations were significantly higher than during the end of the study (August 28–October 9) (Table 3). This is an important factor to consider because O_3 effects on pines and other species are higher in the beginning of the season when stomatal conductance of plants is high and they are more physiologically active (Panek and Goldstein, 2001). In general, our study showed very high O_3 concentrations and exposure indices, indicating a strong potential for negative physiological and biochemical effects on forest trees (Bytnerowicz and Grulke, 1993), predisposing them to drought stress and bark beetle infestation (McBride and Miller, 1999).

Concentrations of HNO_3 and NH_3 in the San Joaquin River drainage were below the levels that could cause direct damage to vegetation (Bytnerowicz et al., 1998). However, due to high deposition velocity of these pollutants (Hanson and Linberg, 1991), even moderately elevated concentrations could contribute substantial amounts of the deposited N to forests, affecting their growth, species composition, as well as surface and ground water quality (Bytnerowicz and Fenn, 1996; Tarnay et al., 2001; Fenn et al., 2003). The calculated maximum dry N deposition for site 1 was 15 kg N ha^{-1} (Table 6). However, it should be noted that annual N deposition values would be substantially higher because our calculations were done for only 4 months and did not include dry deposition of NO_2, particulate NO_3^-, particulate NH_4^+, organic N compounds and wet N deposition. According to our estimates, these other forms of deposited N could increase our predicted values by ~50–60%. Consequently, there is a potential for NO_3^- leaching in this area affecting water quality when compared to NO_3^- leaching Critical Load (CL) of 17 kg N ha^{-1} yr^{-1} (Fenn et al., 2010). At those levels of deposition, there is also a potential for reduction in fine root biomass (Grulke et al., 1998). Serious ecological perturbations in the San Joaquin River drainage are expected even at the calculated minimum dry N deposition values of 3.7–8.2 kg N ha^{-1} (Table 7). There is a potential to initiate (CL = 3.1 kg N ha^{-1} yr^{-1}; Fenn et al., 2008) and cause the shift (CL = 5.2 kg N ha^{-1} yr^{-1}; Fenn et al., 2010) of lichen community functional groups. It is also predicted that at the western end of the transect there is a potential to extirpate lichen species (CL = 10.2 kg N ha^{-1} yr^{-1}; Fenn et al., 2008).

4.5. Evaluation of potential effects on humans and public health implications

Ozone concentrations at the river drainage were elevated above background, especially at locations close to the San Joaquin Valley. These sites experienced similar and greater O_3 concentration than Fresno, a well-known polluted site in the San Joaquin Valley (Cisneros and Perez, 2007). These findings suggest that Shaver Lake and the western sites along the San Joaquin River drainage of this study should be listed in the same category as the city of Fresno, which is considered to be one of the most O_3 polluted urban areas in the country.

The SOMO35 for sites 1–4, 10, and 11 was in the same range (4160–5459 ppb day) as the most polluted sites in Italy (4930–6074 ppb day) reported by Paoletti et al. (2007), and at similar levels as those reported for Northern Europe (Amann et al., 2005). The potential risk to human health in this study could be twice as much as that calculated in Italy because the indices for the Italian sites were calculated for the whole year in contrast to the SOMO35 values in this study were calculated for only 4 months. During the duration of the study, the six sites presented here exceeded the U.S. Federal standard from 42 to 104 days, the California standard from 81 to 108 days, and the European standard from 110 to 114 days. Some sites (1–4) in the San Joaquin River drainage exceeded the U.S, California, and European 8 h health standard almost every day. The European target value directive to protect human health (<25 times a year of concentrations exceeding 50 ppb O_3 as the 8 h mean) was by far surpassed at all sites. These findings clearly show a strong potential for negative O_3 effects on human health in the San Joaquin River drainage and indicate a need to further investigate and inform the public about the risk associated with recreational activities in the southern Sierra Nevada during the summer when O_3 concentrations are the highest. Our results also show that there is need for research on the effects of drainages, canyons, and local topography on O_3 distribution in Sierra Nevada forests in order to inform the public about locations where the risks related to exposures to the pollutant occur. It has been shown that the O_3 exposure regime may greatly vary depending on location in the complex mountain terrain. Locations near ridge tops experience different diurnal patterns than locations near a valley or drainage (Van Ooy and Caroll, 1995; Bytnerowicz, 2005). Locations in or near valleys or drainages usually have a pronounced diurnal distribution of O_3 concentrations, whereas locations near ridge tops have a flatter diurnal pattern. Sites with weaker diurnal variations may experience fewer occurrences of O_3 levels below 40 ppb (Van Ooy and Caroll, 1995). For instance, Bytnerowicz (2005) observed that a western Sierra Nevada site located on a ridge top had a weak diurnal pattern and O_3 concentration in summer with hourly values >40 ppb and 24-h averages >60 ppb. Therefore, concentrations found at that location could potentially have negative effects on human health, especially when considering the new WHO 8-h health guideline set at 50 ppb (WHO, 2003).

5. Conclusions

1. Potential for phytotoxic O_3 effects to sensitive pines and other species are expected since various O_3 exposure indices are among the highest of the studied forested mountain locations in Europe and North America.
2. High O_3 concentrations and meteorological data indicate that polluted air masses from the California Central Valley can deeply penetrate into the Sierra Nevada range and may be transported across to the east side of the Sierra Nevada.
3. Nitric Acid and NH_3 concentrations gradually decreased with distance from the California Central Valley and with increasing elevation due to their higher deposition velocity and reactivity.
4. Concentrations of HNO_3 and NH_3 contributed substantial amounts of N deposition to forests, exceeding the established Critical Loads for nutritional N and having potential effects on forest growth, species composition, as well as surface and ground water quality.

5. Elevated concentrations of O_3, HNO_3, and NH_3 in most of the sites on the San Joaquin River transect in August and September were likely caused by emissions from the McNally fire.
6. High values of various O_3 exposure indices indicate a potentially high risk to human health from this pollutant—most pronounced in the western portion of the San Joaquin River drainage.
7. Understanding of transport patterns and spatial and temporal distribution of air pollutants in the complex topography of the Sierra Nevada are needed for better identification of areas most impacted by air pollution.

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