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# Chapter 16

# Monitoring and modeling of ozone status and effects in the Sierra Nevada: A comparison with studies in North America and Europe

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**Abstract**

Efforts to characterize spatial and temporal distribution of ozone ($O_3$) in the 1999 Sierra Nevada study are compared with similar studies performed in North America and Europe. Statistical models of $O_3$ distribution that were developed based on passive sampler data are discussed in reference to models based on emissions input data. Ozone levels registered during the 1999 Sierra Nevada study are compared with values registered in other mountainous areas, with a special emphasis on the relationship between $O_3$ concentrations and elevation. Reliable information on $O_3$ distribution at a landscape scale is particularly important to detect areas where elevated levels of $O_3$ could be affecting vegetation. For future ecological risk assessment of $O_3$ effects on forests, accurate models of $O_3$ distribution as well as improved understanding of $O_3$ uptake and various factors affecting $O_3$ phytotoxicity are needed.

## 1. Introduction

Air pollution associated with increased industrialization has been recognized as a threat to forest ecosystems for almost 150 years. Many forested areas in Europe and North America have experienced serious environmental problems related to industrial emissions (Wellburn, 1988; Krupa, 1997; Innes and Haron, 2000). In the latter half of the 20th century, deterioration of ponderosa and Jeffrey pine in southern California (Miller et al., 1963), forest decline in the Mediterranean part of Europe (Bussotti and Ferretti, 1998), or dieback of coniferous species in the Mexico City area (Fenn et al., 2002), have been associated with effects of ozone ($O_3$), a secondary air pollutant produced by complex photochemical reactions of nitrogen oxides ($NO_x$) and volatile organic compounds (VOCs) emitted mainly by combustion engines (Seinfeld and Pandis, 1998). Large-scale deterioration of forests may be expected in areas where elevated levels of $O_3$ occur. Trees from heavily polluted areas exhibited reductions in radial growth and higher vulnerability to mortality from insect/disease stresses (Miller et al., 1997). It is predicted that on the western and southern slopes of the Sierra Nevada in California, concentrations of $O_3$ and other photochemical pollutants will increase due to the transport of polluted air from the rapidly developing California Central Valley and the traditionally highly polluted Los Angeles Basin. Negative $O_3$ effects on the Sierra Nevada mixed-conifer forests, especially ponderosa and Jeffrey pines, have been observed for nearly 30 years (Miller and Millecan, 1971; Peterson and Arbaugh, 1992; Carroll et al., Chapter 2, this volume). These observations, however, have not been based on the understanding of large-scale spatial and temporal distribution of $O_3$ concentrations, the effective $O_3$ uptake, physiological and biochemical defensive mechanisms of plants, or interactive effects of $O_3$ stress with other environmental factors such as drought or nitrogen (N) deposition. Better understanding of these factors and complex interactions between them is needed for development of ecologically based models describing responses of forests to air pollution. These models are required for risk assessment of air pollution and recommendations for proper management of Sierra Nevada forests.

This book presents some essential information on natural resources of the Sierra Nevada, spatial and temporal distribution of ozone and other air pollutants, and deposition and uptake of $O_3$ by major forest tree species. The chapters in this book also review the past and present effects of $O_3$ on forests, discuss interactive effects of $O_3$ and N deposition on forests, present methodological needs for effective air pollution monitoring and understanding of the effects of $O_3$ on forests, review management concerns related to air pollution effects on natural resources, and discuss modeling needs for evaluation of deposition and effects of $O_3$ on forests. The results of the Sierra Nevada $O_3$ monitoring program reported in this book come from one of the most extensive and detailed field studies characterizing $O_3$ distribution and effects on forests in complex mountain terrain. In this chapter, we summarize the key findings of the 1999 Sierra Nevada $O_3$ monitoring study and compare them with similar efforts developed in the United States and elsewhere, emphasizing those studies in which passive sampler techniques have been used.

## 2. Applied methodology and models for ozone monitoring in complex terrain

Local $O_3$ concentrations result from different atmospheric physical and chemical processes of formation, transport, deposition, and destruction that depend on meteorological conditions, local topography, and distance to the emission sources of ozone precursors (Zlatev, 1995). The role of complex terrain and associated meteorological processes in $O_3$ distribution in mountainous areas has been studied at several locations, showing that available models of $O_3$ distribution do not reflect pollutant behavior in these areas (Sanz and Millán, 2000). Photochemical air quality models currently used are based on emissions input data, the influence of relevant meteorological parameters, chemical reactions, and dry and wet removal of air pollutants. It has been found that major uncertainties associated with these models are due to model inputs, such as emissions and meteorology, rather than the models (Russell and Dennis, 2000; Tonnesen et al., Chapter 13, this volume). The air quality models based on emissions require extensive spatial and temporal inventories of precursors emissions, and these are usually absent for most of the areas of interest. In addition, the resolution of these models is in the range of 100 km, which is not appropriate for evaluation of biological effects in mountain terrain where spatial variation in $O_3$ concentrations occur in the 10 to 50 km range (Ray, 2001).

The lack of information about air pollutant distribution in complex terrain has increased the interest in developing new techniques to monitor air pollutants that would allow development of landscape-scale models based on monitoring data from dense networks of sites located directly in the receptor areas. Passive samplers allow for this approach because they do not require electric power to operate, are inexpensive and easy to use, and offer the possibility of sufficiently large networks of measurement stations to characterize $O_3$ distribution in the mountainous terrain. Federal laws in the US require monitoring air quality in protected areas to remedy possible existing problems and to prevent significant deterioration in the future. In some of these areas, such as Class I wilderness areas, the absence of electricity is regulatory imposed. Therefore, passive sampling techniques have become extremely useful to examine the spatial and temporal distribution of air pollutant concentrations in these areas and to detect geographical locations at ecological risk. In this sense, additional networks of passive samplers have been deployed to determine baseline and trend concentrations of $O_3$ in protected areas such as national parks and rural areas (Ray, 2001).

The Ogawa $O_3$ passive sampler (Koutrakis et al., 1993) was used in the 1999 Sierra Nevada study, similar to some other efforts recently reported from North America (Ray, 2001; Varns et al., 2001) and Europe (Godzik, 1997; Blum et al., 1997, Sanz et al., 2001; Bytnerowicz et al., 2002a). The Ogawa sampler works on a principle of the controlled passive diffusion of $O_3$ into collecting filter pads and the selective quantitative oxidation of nitrite to nitrate. In the reported study, ambient concentrations of $O_3$ were calculated based on calibration curves developed by direct comparison of passive samplers with collocated active $O_3$ instruments (Lee, Chapter 7; Preisler and Schilling, Chapter 8;

Arbaugh and Bytnerowicz, Chapter 10, this volume). For the $O_3$ distribution models developed for the Sierra Nevada 1999 study, pollutant concentrations from passive samplers and active monitors in the Sierra Nevada and its immediate vicinity, as well as auxiliary meteorological data, were available. The invited modelers could freely decide which modeling approach to apply and data subsets to use. Lee (Chapter 7, this volume) used an elevation-based spatial interpolation method to predict temperature and $O_3$ concentrations over complex terrain to account for the orographic effects on the regionalized variables of interest (Lee and Hogsett, 2001). This modeler used Loess regression to model the spatial variability of $O_3$ as a function of elevation, geographic location, and seasonal mean daily maximum temperature. Preisler and Schilling (Chapter 8, this volume) used a generalized additive regression model to estimate spatial patterns and relationships between predicted $O_3$ exposure and explanatory variables and to predict exposure at non-monitored sites. The fitted model was also used to estimate probability maps for season average ozone levels exceeding critical (or subcritical) levels in the Sierra Nevada region (Arbaugh and Bytnerowicz, Chapter 10, this volume). The explanatory variables—elevation, maximum daily temperature, precipitation, and $O_3$ level at closest active monitor—were significant in the model. Frączek et al. (Chapter 9, this volume) utilized the cokriging geostatistical technique and collateral data (digital elevation model and daily maximum temperatures) to estimate the spatial and temporal distribution of $O_3$ concentrations. These authors applied the Geostatistical Analyst extension to ArcMap 8.1.2 of Environmental Systems Research Institute (ESRI) (Johnston et al., 2001) to generate models of $O_3$ concentration for the study area. In addition, information on $O_3$ and N air pollutants deposition to the western US in summer and winter 1996 was developed with the US Environmental Protection Agency's (EPA) Community Multiscale Air Quality (CMAQ) chemistry-transport model, utilizing emission inventories, meteorological fields, and ambient data (Tonnesen et al., Chapter 13, this volume). There are large uncertainties in the deposition fluxes estimated with that modeling effort, indicating that finer resolution is required to simulate more confidently the wind fields and the spatial variability of deposition fluxes in the complex terrain of the Sierra Nevada.

## 3. Ozone levels in the Sierra Nevada compared with other mountainous areas

Ozone concentrations in the Sierra Nevada varied considerably during 1999, with lowest 2-week averages $\sim 20$ ppb and maximum values $\sim 140$ ppb (Frączek et al., Chapter 9, this volume). In general, there was higher spatial than temporal variability of $O_3$ concentrations. Mean $O_3$ concentrations changed rapidly over short distances as elevation and local topographic settings changed (Lee, Chapter 7, this volume). The areas of highest cumulative ambient $O_3$ were found in the southwestern and west-central Sierra Nevada and in the Lake Tahoe area. High concentrations of $O_3$ also occurred during some periods in the eastern Sierra, especially in the Mammoth Lakes area. The lowest $O_3$ concentrations consistently were found in the northern parts of the range (Frączek et al., Chapter 9; Lee, Chapter 7; Preisler and Schilling, Chapter 8, this volume). Highest $O_3$ levels were observed for the entire range in June, July, and September. In August much lower regional $O_3$ levels were recorded, probably caused by convective air masses and associated periods of rain and down canyon winds (Frączek et al., Chapter 9, this volume). It is likely that changing wind patterns and temperatures along the western and eastern sides of the Sierra Nevada were responsible for the diversity of spatial patterns of $O_3$ distribution. Periods with higher valley temperatures and stronger air flow appeared to coincide with high $O_3$ concentrations along the entire western side of the Sierra Nevada (and a high area along the eastern side), while cooler temperatures resulted in stronger north to south $O_3$ gradients (Frączek et al., Chapter 9, this volume). The CMAQ model for 1996 indicated very high summertime $O_3$ deposition fluxes associated with high concentrations in the San Joaquin Valley and southern portions of the Sierra Nevada (Tonnensen et al., Chapter 13, this volume). Substantially lower rates of $O_3$ deposition were determined during winter because of reduced photochemical production of $O_3$ resulting in low $O_3$ concentrations during that season. A more detailed evaluation of air pollution distribution was performed in summer 1999 in the Sequoia National Park. This study reported elevated $O_3$ concentrations of 41–71 ppb seasonal averages with the highest levels recorded in locations most exposed to the polluted air coming from the California Central Valley (Bytnerowicz et al., 2002b). In summer 2002, a new study is being conducted focusing on distribution of $O_3$ in areas of the Sierra Nevada underrepresented in the 1999 study, such as the eastern regions and air pollution corridors across the Sierra Nevada in the areas of Lake Tahoe, San Joaquin River and Lake Isabella. In addition, a detailed study on distribution of $O_3$ and nitric acid vapor ($HNO_3$) in the Lake Tahoe Basin using passive and active samplers is also being conducted (Alonso et al., unpublished).

Various networks of $O_3$ passive samplers have been deployed in southern California mountains adjacent to the Los Angeles urban area. Special emphasis has been placed on the Class I San Gorgonio wilderness Area in the San Bernardino Mountains (Grosjean et al., 1995; Alonso et al., 2002a). Early studies performed during the 1970s using electronic $O_3$ monitors described a horizontal air pollution gradient along the San Bernardino Mountains with decreasing $O_3$ concentrations from the southwest to the northeast (Miller et al., 1986). Summer season averages in the area ranged from $\sim 30$ ppb $O_3$ in the eastern parts of this mountain range to $\sim 140$ ppb $O_3$ on the western slopes of the San Bernardino Mountains. The long-term study sites established along this horizontal gradient during the 1970s were re-established during the summer 2001 using passive samplers. Passive samplers were deployed in 12 sites throughout the mountains with about 70 km distance from west to east. The results obtained generally agreed with the recent air quality improvement already detected during the last 25 years (Miller et al., 1986). Also, $O_3$ concentrations determined in the San Gorgonio Wilderness of the San Bernardino Mountains in summer 2000 were lower, with seasonal averages between 53 to 59 ppb, and 2-week averages ranging between 37 and 74 ppb (Alonso et al., 2002a). However, summer average $O_3$ concentrations recorded in 2001 reached up to 70 ppb in some locations, indicating that ponderosa and Jeffrey pines may be still threatened by air pollution. Increasing $O_3$ concentrations on the eastern part of the range also seem to be occurring and could be related to the $O_3$ transport from the rapidly developing desert communities of the Palm Springs and Indio area (Alonso et al., 2002b). The combined use of $O_3$ passive samplers with N compound passive samplers provided valuable information about possible interactive effects of $O_3$ and N deposition.

A rural monitoring network of active $O_3$ analyzers is operated by the National Park Service to determine baseline air pollution conditions and trends on a scale of hundreds of km for Class I wilderness areas in the entire US (Ray, 2001). A secondary network of passive sampler monitoring is conducted routinely in 18 national parks, and some passive sampler monitoring has been done in an additional 28 parks, 10 with multiple samplers. The lowest $O_3$ concentrations occur in the northwest and across the northern states of the US, while the highest $O_3$ concentrations are registered in the western US, especially in California (see Web site: http://www2.nature.nps.gov/ard/gas/passives.htm). These findings are consistent with results of pollutant transport from major polluted urban centers in the western US (Tonnesen et al., Chapter 13, this volume). Detailed studies of $O_3$ distribution using passive samplers have been performed in some California national parks. Joshua Tree National Park was used for testing passive samplers because it has high $O_3$ concentrations. Seasonal averages of $O_3$ inside the park were up to 92 ppb in 1999 (see Web site: http://www2.nature.nps.gov/ard/gas/passives.htm). Ozone concentrations registered in the park were higher than averages for the same period recorded in the Los Angeles Basin. Two factors could account for this: overnight titration of $O_3$ by the greater nitric oxide (NO) emissions in the urbanized area, and transport of precursors to the east with $O_3$ formation occurring east of the Los Angeles Basin (Ray, 2001). Multiple passive samplers have been used in Sequoia, Kings Canyon, and Yosemite National Parks where passive samplers have been shown to be useful in studying pollutant distribution in complex terrain. In the eastern US, an extensive air $O_3$ monitoring program in the Smoky National Park with 60 passive samplers that were changed weekly and 4 real-time active monitors was performed in summer 2000, allowing models of $O_3$ distribution with 1-km resolution (see Web site: http://www2.nature.nps.gov/ard/gas/o3study.htm). All those studies showed significantly elevated $O_3$ concentrations in the studied national parks.

The greater Seattle–Tacoma metropolitan region in Washington state is another area where $O_3$ has exceeded the US National Ambient Air Quality Standards (NAAQS) on several occasions in the last 20 years. Ozone forms continuously in the summer and together with its precursors is transported eastward towards the Cascade Mountains and Mount Rainier National Park. A network of passive samplers and electronic monitors was used to measure $O_3$ exposures in four river drainages within the Mount Rainier National Park. It was shown that prevailing northwesterly winds carry $O_3$ precursors that increase $O_3$ levels recorded in the western portion of the park (Brace and Peterson, 1998). Ozone exposure varied considerably over short distances, suggesting that intensive sampling is necessary to quantify spatial patterns of tropospheric $O_3$ in mountainous regions. A more extended network along nine river drainages was deployed to quantify spatial variation in $O_3$ exposure on a regional basis ranging from urban to wildland areas (Cooper and Peterson, 2000). Ozone concentrations were typically higher in rural and protected areas of the Cascade Mountains downwind and east of the urban corridor. Cooper and Peterson (2000) pointed out that $O_3$ distribution in western Washington has a significant regional component with additional variation in magnitude of exposure on a sub-regional basis, depending on spatial variation of meteorology and corresponding transport of $O_3$ and its precursors from urban areas. Because of the regional nature of $O_3$ distribution, it is imperative that regulatory agencies and natural resource managers consider a regional monitoring and management strategy for air quality.

Another study performed further north along the Fraser Valley of British Columbia in Canada confirmed similar $O_3$ distribution patterns, with increasing concentrations from west to east (Runekles and Bowen, 2000). In that study, crop-calibrated passive monitors were used demonstrating that such methods can be used to assess losses to vegetation in regions where ambient $O_3$ levels can only be estimated by atmospheric dispersion models. Where topographic and other features limit the precision of such models, as in regions where little information about ambient $O_3$ exists, the use of calibrated passive monitors can provide a simple means for assessing impact on a range of species *in situ*.

Passive samplers have also been used in the northeastern US where $O_3$ has become the most pervasive phytotoxic air pollutant in wildernes areas. Measurements performed in three wilderness areas in New Hampshire and Vermont showed $O_3$ concentrations between 30 to 45 ppb during the summer (Manning et al., 1996).

For urban areas, the US EPA in collaboration with other local and national agencies manage an extended network of air quality monitoring sites to provide information about $O_3$ concentrations and to warn the public of unhealthy air situations. Similarly, the Canadian National Air Pollution Surveillance Network and the Mexican Metropolitan Networks provide databases for determining air quality in the major urban centers. There are over 4300 monitoring sites operating in North America under these three national networks (Demerjian, 2000). Other efforts using networks of passive samplers have been established to characterize $O_3$ distribution around urban areas in Toronto, Canada (Liu et al., 1995), and Dallas–Fort Worth, Texas (Varns et al., 2001). High $O_3$ concentrations that exceed the 8-h US NAAQS were found at distances greater than 120 km from Dallas city.

In Europe, the need to establish an international cooperative program to develop policies and strategies to control regional air pollution promoted the United Nations/Economic Commission for Europe (UN/ECE) Convention on Long-Range Transboundary Air Pollution (CLRTAP). One of the subsidiary bodies of the CLRTAP is the Environmental Monitoring European Program (EMEP), an instrument to keep and process basic data on air pollution, which is essential for evaluation of the effects of air pollution on forest ecosystems. The combination of known emissions of various pollutants and available monitoring data has enabled the creation of maps along a $150 \times 150$ km grid scale of $O_3$ distribution in Europe. The number of $O_3$ measurement stations in Europe reporting to international coordinating organizations such as EMEP is still relatively small. These stations are also unevenly distributed geographically, with a large number of stations in a few countries and many countries with very few or no $O_3$ data (Simpson, 1996).

EMEP has developed an $O_3$ model capable of addressing both problems of short-term (episodic) and long-term (growing season) $O_3$ concentrations (Simpson, 1993). The model was based upon a combination of previous EMEP activities on photochemical oxidants and the long-term models developed for sulfur and nitrogen pollutants. The new model has been designed with the purpose of simulating $O_3$ formation over long periods of time (1 month to 1 year) over Europe. This model is used to assess the contribution of both anthropogenic and natural emissions to boundary-layer $O_3$ formation and to estimate the likely effect of any control measures on long-term $O_3$ concentrations.

The spatial gradients of $O_3$ produced by the EMEP model are similar to those obtained from the available data in northwestern Europe. There are two main patterns of $O_3$ concentration in Europe: (i) latitudinal, with more frequent episodes of high $O_3$ concentrations in Central Europe (south Germany, Switzerland, northern Italy) than in areas further to the north and west; and (ii) altitudinal, with increased incidents of high $O_3$ concentrations at higher elevations (Grennfelt and Beck, 1994). However, knowledge of the characteristics of $O_3$ regimes over the European region is at best patchy. The model domain covers Europe with a grid resolution that is too large for mountain areas where local conditions significantly modify large-scale patterns. For example, mountains of 1500 m or more in altitude surround much of the Mediterranean. The orography of the region results in site-specific meteorological conditions influenced by sea breezes and upslope winds, which can have marked impacts on patterns of local air quality. Consequently, in the Mediterranean region, the maps produced by EMEP appear to be of less value than elsewhere in Europe, as meso-scale processes result in significant diurnal fluctuations that are not taken into account in the EMEP model (Sanz and Millán, 1998).

Experimental results from the forested areas in Europe indicate that $O_3$ concentrations that may be phytotoxic to forests primarily occur downwind of industrialized areas. In the Swiss Alps, sites of elevations between 410 and 3569 m experienced annual $O_3$ means in 1987 approaching 50 ppb (Wunderli and Gehrig, 1990). In summers 1990–1993, annual $O_3$ mean concentrations in four sites in the Austrian Alps ranging in elevation between 920 and 1758 m were between 26 and 47 ppb (Smidt and Gabler, 1995). In the Sumava and Brdy Mountain ranges in the Czech Republic, 2-week mean concentrations in the summer were approaching 75 ppb (Černy et al., 2002). In rural areas of Lithuania, $O_3$ concentrations typically reach 50 ppb; however, during photochemical episodes, $O_3$ levels might rise even to 85 ppb (Girgzdiene, 1991). Summertime mean peak $O_3$ values in low-elevation Yugoslavian and Greek sites in the 1980s were between 50 and 70 ppb and occurred mainly from noon to early afternoon (Butkovic et al., 1990). In the Madrid Basin in Spain, passive sampler networks have been used to determine areas of possible risk to human and vegetation health (Galán et al., 2001; Sanz et al., 2001). Both studies found higher $O_3$ concentrations in the north and western areas of the Basin, with 2-week averages up to 70 ppb recorded during the summer. Galán et al. (2001) concluded that vegetation growing in areas farther than 20 km from the city might be affected by ozone.

Until recently, little was known about $O_3$ concentrations and its potential phytotoxic effects on the forests of Central and Eastern Europe. During the 1990s, there have been changes in the composition of air pollution in this region (Vancura et al., 2000). The relative role of $O_3$ phytoxicity has increased in this area due to higher local production of $O_3$ precursors from combustion of fossil fuels, long-range transport from western Europe, and recent reductions of sulfur dioxide ($SO_2$) and other industrial emissions. In order to gain a better understanding of the air pollution status in that part of Europe, concentrations of $O_3$, nitrogen dioxide ($NO_2$), and $SO_2$ were monitored with passive samplers aided with a few active monitors during the 1997–1999 growing seasons in 32 forest sites of the Carpathian Mountains (Bytnerowicz et al., 2002a). Highest average hourly $O_3$ concentrations in the western range of the Tatra Mountains reached 100 ppb. The average $O_3$ concentration for all the Carpathian sites during those years ranged between 37 and 44 ppb, with peak 2-week averages reaching 53–61 ppb in the Noveveska Huta site (Slovakia). Elevated levels of the pollutant also characterized other locations throughout the entire Carpathian range, and high spatial diversity of $O_3$ concentration exists, especially in the western Carpathians. Generally, similar spatial patterns of $O_3$ distribution occurred in the Carpathian Mountains during the 3-year study. Clear seasonal patterns in $O_3$ concentration distribution did not occur in the Carpathian range with high $O_3$ concentrations observed both at the beginning and in the middle of the growing season.

## 4. Density of monitoring networks

Previous studies examining the spatial variability of ozone air pollution in the Sierra Nevada found that meteorological processes associated with complex terrain seemed to be more determinant for ozone exposure patterns than the geographical distance from air pollutant sources (Van Ooy and Carroll, 1995). To obtain acceptable resolution for showing these patterns, a dense network of monitoring stations is required. Results of the three models developed for the reported 1999 Sierra Nevada study indicated that a network of 89 $O_3$ passive sampler monitoring sites was sufficiently extensive to cover most of the Sierra Nevada bioregion, with the exception of the southern and southeastern ranges characterized by steep elevation gradients and poor access and a complete lack of continuous $O_3$ monitors. According to the simulation performed with the ESRI Geostatistical Analyst, an increase of the total number of samplers to 124 (for an average of 1 sampler per $\sim 386$ km$^2$ of terrain), adding new sites mostly in the eastern and southern Sierra Nevada, would sufficiently cover the entire Sierra Nevada range, allowing for development of reliable models of $O_3$ distribution (Frączek et al., Chapter 9, this volume). Addition of the passive sampler dataset to the continuous monitoring $O_3$ data from the Sierra Nevada and the surrounding areas significantly decreased the prediction error of spatial patterns of $O_3$ concentrations in the Sierra Nevada (Lee, Chapter 7, this volume) and could significantly improve performance of the applied models.

In the Smoky National Park, a comparison has been made between the 1994 study based on 18 passive sampler monitoring sites and a denser network consisting of 60 passive samplers and 4 real-time active monitors used in 2000. The map of ozone distribution in 1994 showed that the lack of data outside the park caused an under-prediction of $O_3$ on the western area. The network used in summer 2000 with sites ranging from 250 to 2000 m elevation increased the level of detail in understanding $O_3$ distribution on ridges and mountains peaks that were previously underrepresented and allowed for the development of models at 1-km resolution (see Web site: http://www2.nature.nps.gov/ard/gas/passives.htm).

A simulation exercise was performed with the ESRI Geostatistical Analyst for the Carpathian Mountains study to determine how many points are needed to provide satisfactory predictions of ozone distribution for the entire area of the monitored range (Bytnerowicz et al., 2002a). In general, the simulation exercise indicated that if the number of monitoring sites increased from the original 32 to 140 (1 site per 1000 $km^2$), satisfactory confidence in predicting $O_3$ concentrations for the Carpathian Mountains range would cover about 99% of the entire forested area (Bytnerowicz et al., 2002a).

## 5. Effects of elevation on ozone distribution

Increases of $O_3$ concentrations with elevation have been reported in different mountain locations (Gabler et al., 1990; Monn et al., 1990; Proyou et al., 1991; Loibl et al., 1994; Smidt and Gabler, 1995; Matyssek et al., 1997; Rennenberg et al., 1997). Similarly, ambient $O_3$ continuously increased with elevation up to 2100 m in the Cascade Mountains (Washington state) on three of the four transects analyzed (Brace and Peterson, 1998). High ozone levels recorded at higher altitude sites have been related to an increase in the $O_3$ background level due to the accumulation of photochemically produced $O_3$ at a wider (regional and synoptic) scale and to the redistribution and exchange processes between the boundary layer and the free troposphere (Gerosa et al., 1999).

However, on three profiles studied in the Bavarian and Austrian Alps in the 1980s, effects of elevation change on $O_3$ distribution patterns were not reported: $O_3$ levels increased with elevation up to about 1700–1800 m, then with additional gain in elevation (up to 3000 m) an increase of $O_3$ concentrations was not observed (Puxbaum et al., 1991). Similar patterns have been described in the San Bernardino Mountains in southern California. Miller et al. (1986) found increasing $O_3$ concentrations with elevation when comparing valley locations (400 m) with mountain areas at around 1800 m. While the $O_3$ concentrations during daylight hours were not significantly different in the mountain and valley locations, the nighttime concentrations were distinctly higher at the mountain locations. When examining vertical distribution of $O_3$ more closely with passive sampler data on a gradient from 1200 to 2700 m, a clear effect of elevation change on $O_3$ concentrations was not found (Alonso et al., 2002a). Similar findings were reported from an air pollution monitoring campaign using active monitors and passive samplers in the Sequoia National Park in the western Sierra Nevada in summer 1999. With increasing elevation and distance from the pollution source area (California Central Valley), concentrations of $O_3$, ammonia ($NH_3$), and $HNO_3$ decreased (Bytnerowicz et al., 2002b). In the Smoky National Park, a strong increase of $O_3$ concentrations with elevation up to 2000 m was determined in summer 2000 in the south side of the Park using passive samplers (see Web site: http://www2.nature.nps.gov/ard/gas/passives.htm). In sites on the north side of the park, although showing a similar trend, the relationship between $O_3$ and elevation was not significant. In this case, sites were not up a single ridge or drainage; thus, other spatial variation can complicate the analysis. In Yosemite National Park, two patterns relating ozone to elevation could be found. Sites showing a clear increase of $O_3$ with elevation up to about 2000 m could be more influenced by upslope flows that bring $O_3$ from the San Joaquin Valley. Sites above this elevation did not show a consistent gradient with elevation being influenced mostly by free tropospheric air that is less directly connected with a specific source or polluted area (Ray, 2001).

In the 1999 Sierra Nevada study, increasing elevation was positively associated with increasing ambient $O_3$ concentrations in a residual analysis conducted in one model (Preisler and Schilling, Chapter 8, this volume), while another analysis showed a leveling of seasonal mean ambient $O_3$ when elevation exceeded 1500 m (Lee, Chapter 7, this volume). These results may not be contradictory. Ambient $O_3$ may tend to increase with elevation; however, at higher elevations, temperature decreases and therefore the potential for $O_3$ formation is lower. Both factors together may lead to the observation of a leveling of ambient $O_3$ concentrations. In addition, after initial increases of $O_3$ concentrations caused by reactions of VOCs and $NO_x$ downwind of the photochemical smog source areas, decline of concentrations takes place due to dilution of the pollutant, its uptake by vegetation, and reactions with various landscape features.

In the Carpathian study in 1997–1999, a relationship between elevation and $O_3$ concentrations was not observed on the north-west/south-east transect in the Morava–Silesian Mountains. However, an increase of $O_3$ concentrations with elevation was clear on the south-east/north-west transect in the same area; for the 1997 and 1998 seasons the increase was logarithmic, and for the 1999 season linear. On the basis of this evidence, elevation could be used as a covariate for estimation of $O_3$ concentration distribution in this area (Bytnerowicz et al., 1999).

## 6. Effects of ozone on forest health

Since the identification of the unique injury symptoms caused by ozone on conifers, researchers have conducted field surveys for quantifying and monitoring the effects of air pollution on forest ecosystems. Foliar chlorotic mottle and foliar retention are the main attributes used to assess ozone injury. A variety of survey approaches and indices of ozone injury have been used by different agencies in the US (Duriscoe et al., 1996). Similarly, several ozone exposure indices have been used as indicators of biological injury (Lefohn, 1992). Previous studies found linear relationships between ambient $O_3$ exposure and visible foliar injury in the western Sierra Nevada (Miller et al., 1996; Salardino, 1996; Arbaugh et al., 1998). The Ozone Injury Index (OII) was useful to measure cumulative crown injury caused by ozone and was significantly correlated to 4-year summer cumulative 24-h ambient $O_3$ exposure indices, especially with SUM0 (cumulative sum of all hourly $O_3$ concentrations over an exposure period) (Arbaugh et al., 1998). Analyses showed that OII is functionally equivalent to the Forest Pest Management (FPM) index used in other approaches (Arbaugh et al., 1998). In the 1999 Sierra Nevada study, high $O_3$ exposure areas were not always associated to moderate or high injury to pines assessed using the FPM index (Arbaugh and Bytnerowicz, Chapter 10, this volume). There was a relationship between ozone exposure and injury index only when average seasonal ambient $O_3$ exceeded 60 ppb. The FPM index evaluates long-term effects of $O_3$ exposure since it is quantified by noting the youngest whorl of needles showing chlorotic mottle symptoms from ozone. In this sense, FPM index will be more related to $O_3$ exposure accumulated over several years than to the single year exposure calculated in this study.

Site-specific factors, such as aspect, soil water balance, and phenotypic response by local populations, also affect expression of visible injury (Arbaugh et al., 1998; Grulke, 1999). Effects of $O_3$ on vegetation are more closely related to the $O_3$ dose absorbed through the stomata than to $O_3$ exposure in the atmosphere (Musselman and Massman, 1999). This complexity of the plant responses to $O_3$ exposure have resulted in the shift from focusing on exposure-response relationships towards plant $O_3$ uptake or flux models that can better estimate the damaging component of $O_3$ on vegetation. High ambient $O_3$ concentrations are not always coupled to high $O_3$ uptake by trees. Goldstein et al. (Chapter 4) and Panek et al. (Chapter 14, this volume) have shown experimentally the disparity between $O_3$ uptake and $O_3$ concentrations in the pine forests of the Sierra Nevada. Different biotic and abiotic factors control $O_3$ uptake and must be included in models of $O_3$ deposition (Grulke, Chapter 3; Panek et al. Chapter 14, this volume). On a regional basis $O_3$ deposition to ecosystems occur not only through stomatal uptake but also through non-stomatal deposition or through gas phase chemical reaction in the forest canopy (Goldstein et al., Chapter 4; Panek et al., Chapter 14, this volume). Estimations of $O_3$ deposition are needed at different scales from the leaf level to the whole ecosystem to evaluate the threat of air pollution to forest ecosystems. Quantifying total $O_3$

deposition is important to understand the temporal and spatial distribution of $O_3$ concentrations.

A similar evolution from exposure-response to pollutant uptake approach has occurred in Europe under the UN/ECE CLRTAP as an internationally legally binding instrument to deal with problems of air pollution on a broad regional basis. The CLRTAP has adopted the methodology of critical levels to develop policies and strategies to control regional air pollution. Critical levels of $O_3$ are defined as concentration ranges above which negative effects on vegetation can be expected (Skärby et al., 1998). In defining critical levels, there are two major approaches: Level I and Level II. The Level I was established to protect the most sensitive receptor based on exposure-response relationships. A Level II approach is now being developed to take into account the response of different species and modifying factors, such as the presence of other pollutants, vapor pressure deficit, soil moisture content, influence of mineral nutrition, etc., that affect plant response to ozone. Models of stomatal flux of $O_3$ have been developed to estimate regional-scale $O_3$ uptake across Europe (Emberson et al., 2000).

The potential phytotoxicty of $O_3$ is not only influenced by stomatal conductance and $O_3$ uptake but also by the ability of the plant to activate both protective and reparation processes. These processes are also influenced by environmental factors, plant phenology, physiological status, and tree age. Grulke (Chapter 3, this volume) gives some recommendations to improve $O_3$ injury field assessments that consider the interaction of these modifying factors. These suggestions have great value for developing long-term foliar monitoring networks for the Sierra Nevada (Plymale et al., Chapter 12, this volume).

## 7. Future directions for ozone risk assessment

Efforts for $O_3$ risk assessment to forest ecosystems for the protection of ecological, economic, and aesthetic values of vegetation need to quantify $O_3$ exposure. Both spatial and temporal distribution of $O_3$ concentrations and pollutant uptake by vegetation should be considered on a regional scale, including complex terrain areas where $O_3$ monitoring is limited. The use of passive sampler systems have allowed the development of statistical models describing patterns of ambient $O_3$ over space and time in the Sierra Nevada. Also the combine use of $O_3$ and N compounds passive samplers will offer promising information on the interactive effects of multiple pollutants. Available models of $O_3$ deposition and uptake also offer the possibility to estimate the absorbed dose of pollutants that would be affecting vegetation. However, the most widely used exposure indices for deposition models, such as SUM0 in the US and AOT40 in Europe, accumulate hourly $O_3$ concentrations over certain periods, resulting in different weights to high and low $O_3$ concentrations. Calculation of those indices required hourly values from continuous $O_3$ analyzers operating for long periods of time. Unfortunately, forests located in mountain areas are not well covered by networks of $O_3$ monitors. Promising efforts are being developed by Mazzali et al. (2002) and Tuovinen (2002) to use passive sampler data to calculate cumulative $O_3$ exposure indices to assess potential damage to vegetation.

The development of accurate models for estimating $O_3$ distribution and uptake by vegetation is one of the main challenges for $O_3$ risk assessment in the Sierra Nevada. Improving the design and performance of foliar surveys is also needed to assess the impact of ozone. Moreover, the possibility to use other indicators of changes in forest ecosystems should be considered. Cooperation in research combining and comparing different approaches is central to cope with the problem of air pollution, which does not recognize national boundaries.

Passive samplers will provide valuable information for a better understanding of air pollution distribution in remote areas where traditional electronic equipment cannot be used. Results of these monitoring efforts will help in identifying these areas where potential problems may be occurring and where more detailed evaluation of environmental status should be performed. The purpose of this book has been to provide new knowledge on natural resources and the effects of air pollution on the Sierra Nevada forests that will help in future efforts to preserve healthy air quality for forests in these magnificent mountains.

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