Elevational and spatial variation in daytime ozone concentrations in the Virginia Blue Ridge Mountains: implications for forest exposure

F. S. Gilliam, J. T. Sigmon, M. A. Reiter, and D. O. Krovitz

Department of Environmental Sciences, Clark Hall, University of Virginia, Charlottesville, VA 22903, U.S.A.

Received February 15, 1988
Accepted November 16, 1988


Ozone (O$_3$) concentrations before and during the early growing season were monitored continuously at two closely located areas in the Virginia Blue Ridge Mountains: a deciduous forest watershed (Shaver Hollow) and a predominantly open grassy meadow (Big Meadows). In addition, O$_3$ concentrations and canopy development (percent leaf emergence) were measured simultaneously at three sites along the elevational gradient of the Shaver Hollow watershed. When the canopy at Shaver Hollow was leafless, patterns of O$_3$ concentration were similar between the forest and meadow, increasing steadily toward early spring, with low variability of hourly daytime measurements. Greater differences in O$_3$ concentration between forest and meadow and higher variability occurred during the period May–June, a time when the canopy changed from 15 to 100% leaf emergence. Several factors varying along the elevational gradient, related to both canopy structure and meteorological conditions, appeared to be important in affecting absolute O$_3$ concentrations as well as the variability of daytime means in Shaver Hollow. The high metabolic activity of new leaves may act as a sink for O$_3$; the canopy itself may act as a physical barrier to O$_3$ transport, restricting mixing of O$_3$ to intermittent bursts. The coincidence of seasonal increases in O$_3$ concentration in late spring and early summer with the development of new leaves of dominant deciduous trees in this watershed suggests that O$_3$ may represent a potential problem for eastern hardwood forests.


Les concentrations d’ozone (O$_3$) avant et pendant le début de la saison de croissance ont été enregistrées de façon continue à deux endroits rapprochés dans les montagnes du Blue Ridge en Virginie : un bassin hydrographique formé de forêts décidues (Shaver Hollow) et une prairie herbeuse plutôt ouverte (Big Meadows). En outre, les concentrations de O$_3$ et le développement du couvert foliaire (pourcentage d’emergence foliaire) ont été mesurés simultanément à trois endroits le long du gradient altitudinal du bassin hydrographique de Shaver Hollow. Au moment où le couvert foliaire du Shaver Hollow était non feuillé, les variations de la concentration de O$_3$ étaient semblables entre la forêt et la prairie, et elles augmentaient régulièrement au début du printemps en offrant une faible variabilité durant les mesures horaires effectuées de jour. Des différences plus considérables de la concentration de O$_3$ entre la forêt et la prairie et une plus grande variabilité sont survenues durant les mois de mai et juin, au moment où le couvert foliaire évoluait d’une émergence foliaire passant de 15 à 100%. Plusieurs facteurs qui variaient le long du gradient altitudinal, reliés aussi bien à la structure du couvert foliaire qu’aux conditions météorologiques, ont semblé importants pour leur effet sur les niveaux absolus de O$_3$, de même que sur la variabilité des moyennes de jour dans le Shaver Hollow. La forte activité métabolique des nouvelles feuilles pourrait agir comme un réservoir pour O$_3$ ; le couvert foliaire lui-même peut constituer un obstacle au déploiement de O$_3$, restreignant le mélange de O$_3$ à des poussées intermittentes. La coïncidence des augmentations saisonnières des concentrations de O$_3$ vers la fin du printemps et au début de l’été avec le développement des nouvelles feuilles des sujets décidus dominants dans ce bassin hydrographique indique que O$_3$ pourrait présenter un problème potentiel pour la forêt décidue de l’Est américain.

[Traduit par la revue]

Introduction

Ozone (O$_3$) has been cited as an important component of a complex of atmospheric pollutants that may have adverse effects on forest ecosystems in both Europe and North America (Ashmore et al. 1985; Linzon 1986). As a result of oxidative foliar damage, O$_3$ can reduce photosynthetic rates (and subsequently decrease growth and yield) in crops and forest tree species (Reich and Amundson 1985). Chappelka and Chevone (1986) showed that interaction of simulated acid rain and O$_3$ at intermediate concentrations (50 and 100 ppb) caused significant reductions of several growth parameters in white ash. It has been hypothesized that O$_3$ may increase the leachability of certain ions from deciduous leaf tissues, particularly in response to increased acid deposition (Skeffington and Roberts 1985; Chappelka and Chevone 1986).

In addition to being a chronic pollutant in urban areas (Brimblecombe 1986), O$_3$ is a problem in rural areas, where high concentrations appear to be the result of a variety of factors, especially long-distance transport of urban plumes within slowly moving high-pressure systems (Altschuller 1986). Meagher et al. (1987) reported long-term O$_3$ monitoring data for several rural sites in the southeastern United States, indicating that rural O$_3$ concentrations equaled, and sometimes exceeded, urban O$_3$ concentrations in the same region. Furthermore, Linzon (1986), reviewing studies of the effects of gaseous pollutants on forests in eastern North America, concluded that O$_3$ is the dominant phytotoxic air pollutant in this region.

There is great variability in sensitivity to O$_3$ damage...
among forest tree species, with many important deciduous species considered to be moderately or highly sensitive (Linzon 1986). Studies have also shown that concentrations of O$_3$ at lower elevations exhibit greater diurnal variation than those at higher elevations (Broder and Gygax 1985). Thus, O$_3$ stress upon deciduous forests is potentially affected by both the species composition and the elevation of the forest.

We examined seasonal changes in O$_3$ concentration at two closely located sites in the Blue Ridge Mountains of northern Virginia. The data comparisons presented here address hypotheses concerning the turbulent transfer of O$_3$ to forests and the effects of a hardwood forest canopy (and its phenological development) on O$_3$ concentrations within the canopy. Further emphasis is placed on the interaction of O$_3$ and canopy development with respect to the elevational gradient of the watershed.

Most recent field studies on O$_3$ concentrations in the lower troposphere have been on a larger, regional scale (e.g., Wolff and Liyo 1980; Meagher et al. 1987) or at single locations (e.g., Droppo 1985), and few studies have examined patterns of O$_3$ concentration with respect to elevational changes on a subregional scale. It is on this scale, however, that changes in atmospheric O$_3$ may be more meaningful in terms of forest exposure. The steep slopes of watersheds in the Blue Ridge Mountains impose elevational gradients on many environmental variables (e.g., solar radiation, temperature, wind speed). These variables, in turn, interact with elevational aspects of the forest canopy (e.g., canopy height and phenological changes). The objectives of this paper are to compare seasonal O$_3$ patterns between a forest site and a nearby meadow site and to examine O$_3$ conditions inside a hardwood forest canopy as they vary along an elevational gradient.

**Methods**

**Study site**

Data were collected at the Shaver Hollow watershed and Big Meadows Station located in the north central section of Shenandoah National Park, Virginia. Shaver Hollow covers approximately 223 ha and has a predominantly west to northwest aspect. The area of the watershed used for this study ranges from 503 to 1037 m in elevation, with slopes averaging between 50 and 60%. The forest is second-growth mixed hardwood, with chestnut oak (Quercus prinus L.) and northern red oak (Q. rubra L.) being codominant. Big Meadows Station, operated by Shenandoah National Park, is a predominantly open grassy area 15 km south of Shaver Hollow. For a description of Big Meadows see Duchelle et al. (1983).

**Field design and instrumentation**

At the Shaver Hollow watershed, three micrometeorological towers with associated instrument shelters were established at elevations of 1014 m (site 1: tower 16.5 m, canopy 13.7 m), 716 m (site 2: tower 21.9 m, canopy 13.7 m), and 524 m (site 3: tower 27.4 m, canopy 22.9 m). No trees were removed during tower construction, therefore the canopy was not altered. Wind speed and direction, ambient temperature, relative humidity, and solar radiation were measured at the top of each tower (above the canopy). Instruments were sampled at 5-s intervals, using a Campbell Scientific 21XL Datalogger in the instrument shelter. All signals were stored on cassette tape, as 15-min and 1-h averages, for transportation and analysis.

Ozone concentrations at each Shaver Hollow site were measured within the canopy at a height of 3 m above the forest floor, using Thermo Electron Model 49/103 uv photometric ozone analyzers. The analyzers were sampled at 5-s intervals by the datalogger, and 15-min and 1-h averages were computed and stored on cassette tape, along with the basic climatic data. Ozone concentrations at Big Meadows were measured in the open meadow at a height of 4 m, with a Dasibi Model 1003 RS uv photometric ozone analyzer. Hourly averages were obtained from Shenandoah National Park. Ozone concentrations presented in this paper represent daily averages of hourly daytime (08:00-18:00 EST) values.

Multipoint calibrations of the O$_3$ analyzers (Shaver Hollow) against a Columbia Scientific Instruments primary standard were performed on 7 May 1987 at site 1, 14 May 1987 at site 2, and 12 May 1987 at site 3. On the basis of these calibrations, data collected at Shaver Hollow have an accuracy of ±5%. Data collected before these calibrations were mathematically corrected for elevation by multiplying hourly average O$_3$ concentrations by the ratio of ambient barometric pressure to sea-level pressure. Once calibrations were completed, all corrections for elevation were performed internally by the analyzer. All data from Big Meadows were within ±5% and in compliance with both State of Virginia and U.S. Environmental Protection Agency guidelines (J. Watkins, personal communication).

Canopy phenology was characterized as percent leaf emergence. This was estimated periodically from canopy photographs and field observations throughout the early growing season.

**Results**

Figure 1 shows O$_3$ concentrations at Shaver Hollow site 1 and at Big Meadows for February–March, along with corresponding standard deviations about daily means. Concentration patterns were similar between the two sites, with daily averages slightly higher at Big Meadows. Maximum values in this period occurred during a 2-day peak in early March (60 and 67 ppb at site 1 and Big Meadows, respectively). Minimum values occurred at the start of the period (20 and 30 ppb, respectively). Also shown in Fig. 1 are regression lines generated from 7-d running averages of O$_3$ concentration over this period. Daytime O$_3$ concentrations at both sites appeared to increase by about 1.6 ppb O$_3$/week from February through March. Standard deviations were low at both sites throughout the period.

Ozone concentrations and standard deviations for both sites from May through June are presented in Fig. 2. Once again, patterns were strikingly similar between sites. However, the differences in mean daily values between Big Meadows and site 1 tended to be greater during this period than in the February–March period, particularly beginning...
FIG. 2. Daytime O₃ concentrations (ppb) for site 1 at Shaver Hollow (solid line indicates means, crosses indicate standard deviations) and Big Meadows (dotted line indicates means, diamonds indicate standard deviations) from 4 May to 19 June 1987. Straight lines represent regressions of 7-d running averages on Julian day.

FIG. 3. Mean daytime O₃ concentrations (ppb) and standard deviations (●) for sites 1, 2, and 3 at Shaver Hollow, from 20 April to 19 June 1987.

in late May. Maximum values of 79 and 84 ppb at site 1 and Big Meadows, respectively, occurred towards mid-May. Minimum values (6 and 19 ppb, respectively) occurred in late May. In contrast to those in February-March (Fig. 1), O₃ concentrations in May–June exhibited neither increasing nor decreasing trends, indicated by zero slopes of the regression lines for 7-d running averages (Fig. 2). Daily variation in O₃ concentrations (standard deviation) was much greater during May–June than during February–March.

Daytime O₃ concentrations for all three Shaver Hollow sites from April through June are shown in Fig. 3. April–June was chosen because the leaves of the deciduous trees at the Shaver Hollow sites are developing over this period. Figure 4 shows seasonal changes in percent canopy leaf emergence at the three sites. Canopy development followed the elevational gradient of the watershed, with 50% full canopy occurring in approximately early, mid, and late May for sites 3, 2, and 1, respectively.

Throughout the period April–June, day to day fluctuations in O₃ concentration were very similar among the three sites at Shaver Hollow. There was no pattern of either increasing or decreasing concentrations during this period. Although mean O₃ values for the sites were identical on many occasions, large differences between sites did occur (e.g., days 114 and 130, Fig. 3). Standard deviations about the daily means were smallest at site 1. Increases in standard deviation appeared to coincide with canopy development at all sites. Patterns of mean daily wind speed were similar among the three sites at Shaver Hollow (Fig. 5). However, wind speeds were highest and most variable at site 1, followed by sites 2 and 3, apparently in response to the elevational gradient of the watershed.

**Discussion**

The transfer of mass (including O₃) from the atmosphere to the forest canopy can be highly intermittent because of the intermittency of the transporting eddies (Denmead and Bradley 1985). The canopy itself may act as a sink for O₃ through diffusion into stomatal cavities and subsequent reaction with the surface of leaf mesophyll cells (Townsend 1974), so that under similar ambient conditions, differences in mean O₃ concentration are expected to occur between an open site and a within-canopy site. Furthermore, for deciduous forests, the degree of these effects should vary with the phenological development of the canopy. Therefore, the effect of the canopy on O₃ is expected to be lower concentrations and larger standard deviations within the canopy compared with nearby open areas.

There are other factors which can affect O₃ concentrations that are independent of phenological changes. For grassland, Colbeck and Harrison (1985) showed that differences in the 24-h maximum and minimum O₃ concentrations were largest in the summer and smallest in the winter. They concluded that changes in diurnal variation resulted from seasonal changes in meteorological conditions which affected the downward mixing of O₃, the photochemical
production and transport of O$_3$, and the formation of nocturnal inversions.

Meteorological factors have also been shown to influence the variation in diurnal O$_3$ with respect to elevation in heterogeneous terrain (Kelly et al. 1984; Broder and Gygax 1985). Higher diurnal O$_3$ variability may be expected at lower elevations as a result of differences between mountain and valley areas in wind speed (mixing processes), photochemical O$_3$ production (precursor availability), and differences in deposition rates due to development of inversion layers.

By considering only daytime values, differences in O$_3$ concentrations induced by diurnal processes were reduced. Because the sites are near one another, it was felt that differences among the sites would then be due primarily to differences in canopy structure and (or) local mixing processes.

**Forest site versus meadow site**

The potential effects of a deciduous canopy upon O$_3$ concentrations within that canopy were investigated by comparing O$_3$ concentrations at site 1 with those at Big Meadows Station. Because these sites are only 15 km apart and differ in elevation by only 20 m, it is reasonable to assume that their macroclimates are similar. Differences in O$_3$ concentrations between the sites, therefore, may be attributed largely to the presence of the forest canopy at site 1.

During winter, when the canopy was leafless, similar patterns of daytime O$_3$ concentrations were observed between sites (Fig. 1). Mean O$_3$ values tended to be slightly higher at Big Meadows, supporting the hypothesis that the presence of even a leafless canopy may have some effect on ambient O$_3$ concentrations, as suggested by Smith (1981).

There was a definite pattern of increasing O$_3$ concentrations from the beginning of February to the end of March (Fig. 1), consistent with the seasonal O$_3$ pattern found for rural sites by Meagher et al. (1987). As there is no apparent source of hydrocarbon precursors within the study area during this leafless period, the increases in O$_3$ were probably a result of atmospheric transport from urban areas. Other studies have shown that long-distance transport is a major source of O$_3$ in rural areas (Coffey and Stasiuk 1975; Alshuller 1986).

Greater differences in O$_3$ concentrations between site 1 and Big Meadows occurred in the period May–June (Fig. 2). These differences generally increased toward June, closely following canopy development at site 1. Trees at this site were approximately 50% full canopy in mid-May and full canopy in early June (Fig. 4), supporting the hypothesis that new leaves can act as a sink for O$_3$ (Townsend 1974; Smith 1981). The greater percent leaf emergence at site 3 compared with the other two sites (Fig. 3) also tended to exhibit less variation in daytime O$_3$ concentrations than did site 3 (Fig. 3). One interpretation of these data is that the mixing of O$_3$ through the canopy is more uniform at sites 1 and 2 than at site 3. The more exposed locations and less developed canopies of sites 1 and 2 could facilitate a more uniform downward mixing of O$_3$ within the canopy. The more complete canopy of site 3, combined with lower wind speeds, would favor an intermittent mixing of O$_3$. The greater percent leaf emergence at site 3 compared with the other two sites (Fig. 4) might also play a role by providing a larger active sink for O$_3$ than that available at the same times for sites 1 and 2.

Data presented here are also consistent with elevational and seasonal variations in meteorological factors affecting O$_3$ concentrations which are largely independent of canopy phenology. Broder and Gygax (1985) attributed decreases in the diurnal variability of O$_3$ concentrations with increasing elevation to enhanced depletion at lower elevations under nighttime inversions. Colbeck and Harrison (1985) demonstrated that diurnal variation in O$_3$ concentrations was substantial in the summer and minimal in the winter, again by comparing the three sites in Shaver Hollow. Canopy development followed the elevational gradient of the watershed, with 50% leaf emergence occurring at sites 1, 2, and 3 on or around Julian days 139, 132, and 126, respectively (Fig. 4). These days coincide with noticeable increases in the average standard deviations of daytime O$_3$ concentrations at the respective sites (Fig. 3).

Our data show that the forest canopy at site 1 is nearly fully developed at a time when daytime O$_3$ concentrations within the canopy have increased from approximately 35 ppb (no canopy development, Fig. 1) to often in excess of 60–70 ppb (Fig. 2). The sensitivity of tree leaves to O$_3$ can vary with leaf growth and development, with underdeveloped leaves generally more sensitive than fully developed leaves (Townsend and Dochinger 1974; National Academy of Sciences 1977). Thus, the coincidence of seasonal increases in O$_3$ concentrations with the development of deciduous forest canopies may be important in determining overall O$_3$ effects on the forest. It should be noted that the O$_3$ concentrations reported here are approximately within the range of values considered from laboratory and field studies to be potentially harmful to forests (Reich and Amundson 1985; Linzon 1986).

There are both canopy-related and meteorological explanations for the increases in variability of standard deviations observed along the elevational gradient. For example, the developing leaves may act as sinks for O$_3$ within the canopy, particularly during daylight hours, when stomata are open (Townsend 1974). The leaves may also form a physical barrier which restricts the mixing of O$_3$ through the canopy to intermittent bursts (Denmead and Bradley 1985). Whereas the amount of O$_3$ removed by the leaves should vary with their daily metabolic activity (Townsend and Dochinger 1974), the barrier effect of the leaves might be seasonal and correlated with local wind speed, turbulence, and canopy thickness. Thus, the presence of a leafy canopy may simultaneously dampen variation by providing a sink for O$_3$ and increase variation by creating a barrier favoring intermittent mixing of O$_3$ downward through the canopy.

Sites 1 and 2 generally experienced higher and more variable wind speeds than site 3 during the period April–June (Fig. 5). Coincidentally, sites 1 and 2 also tended to exhibit less variation in daytime O$_3$ concentrations than did site 3 (Fig. 3). One interpretation of these data is that the mixing of O$_3$ through the canopy is more uniform at sites 1 and 2 than at site 3. The more exposed locations and less developed canopies of sites 1 and 2 could facilitate a more uniform downward mixing of O$_3$ within the canopy. The more complete canopy of site 3, combined with lower wind speeds, would favor an intermittent mixing of O$_3$. The greater percent leaf emergence at site 3 compared with the other two sites (Fig. 4) might also play a role by providing a larger active sink for O$_3$ than that available at the same times for sites 1 and 2.

Data presented here are also consistent with elevational and seasonal variations in meteorological factors affecting O$_3$ concentrations which are largely independent of canopy phenology. Broder and Gygax (1985) attributed decreases in the diurnal variability of O$_3$ concentrations with increasing elevation to enhanced depletion at lower elevations under nighttime inversions. Colbeck and Harrison (1985) demonstrated that diurnal variation in O$_3$ concentrations was substantial in the summer and minimal in the winter, again...
as a result of enhanced nighttime depletion related to stability effects. Differences in \( \text{O}_3 \) variability between winter and spring at Shaver Hollow (cf. Figs. 1 and 2) are consistent with these results. As stated earlier, the use of daytime values and the close proximity of the three watershed sites should minimize the variability caused by enhanced nighttime depletion. It is likely, however, that meteorological and canopy effects are confounded in field studies.

While most investigations of the effect of \( \text{O}_3 \) on trees have dealt with coniferous species (National Academy of Sciences 1977), several important deciduous forest species, including tulip poplar, sweetgum, and several oaks, have been shown to be susceptible to \( \text{O}_3 \) damage (Davis and Wilhour 1976). Data presented in this paper show that maximum \( \text{O}_3 \) concentrations occur at Shaver Hollow when leaves of the overstory canopy are nearly fully developed. This is a time when leaves are most sensitive to \( \text{O}_3 \) damage (Townsend and Dochinger 1974). Furthermore, \( \text{O}_3 \) concentrations at Shaver Hollow are similar to those at other rural forest sites (Coffey and Stasiuk 1975; Meagher et al. 1987) and are within the range considered from both laboratory and field studies to be potentially harmful to forests (Reich and Amundson 1985; Linzon 1986). It is therefore conceivable that deciduous forests of the eastern United States may be experiencing \( \text{O}_3 \) stress. Because many park and commercial forests have significant deciduous components, it seems important to study the effect of \( \text{O}_3 \) upon natural assemblages of these deciduous species.

Acknowledgements

This work was supported by the Environmental Protection Agency through a cooperative agreement with the University of Virginia, and through a cooperative agreement forming part of the Mountain Cloud Chemistry Project (Dr. Volker A. Mohren, Principal Investigator). This work was also part of the Shenandoah Watershed Study. Although the research was funded by the United States Environmental Protection Agency, this paper has not been subjected to agency review, therefore it does not necessarily reflect the views of the agency and no official endorsement should be inferred. The authors wish to thank Dr. Ronald Bradow, Dr. James Meagher, and anonymous reviewers for their comments on the manuscript, and the U.S. Department of the Interior, National Park Service, and Jim Watkins for providing data from the Big Meadows site. Field assistance was provided by Wendy McIntyre.


Ashmore, M., Bell, N., and Rutter, J. 1985. The role of ozone in forest damage in West Germany. Ambio, 14: 81-87.