

PROTECTING AGRICULTURAL CROPS FROM OZONE EXPOSURES KEY ISSUES AND FUTURE RESEARCH DIRECTIONS

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EXECUTIVE SUMMARY

The ubiquity and toxicity of ambient air O_3 is well documented. Because O_3 is an omnipresent air pollutant that affects both human health and vegetation, the U.S. Environmental Protection Agency (EPA) has established both primary and secondary standards. There is no requirement that the primary and secondary standards be identical, nor is there any requirement that only a single expression of the standard be used (i.e., an average concentration for a single time period versus multiple exceedances or integrated exposures). Any effort to propose a secondary standard, whose form is different than the current form of the primary and secondary standard, implies that either (1) the current form is inappropriate for protecting the public welfare or (2) a more restrictive value of the current form of the standard is required.

There have been indications reported in the literature that the current form of the standard may not be appropriate for protecting vegetation from O_3 exposures. The purpose of this report is to identify and review some of the key issues related to assessing the effects of O_3 on vegetation. Our report has reviewed the available information on (1) components of O_3 exposure that elicit adverse effects on vegetation, (2) ways to describe these components in the form of O_3 exposure indices that may be useful in the standard-setting process for protecting vegetation, (3) the change in nonattainment status that may occur should the existing O_3 standard be modified, and (4) the need for future research efforts to explore the development of a multi-parameter index to protect vegetation from O_3 exposure.

Our results, using a select set of National Crop Loss Assessment Network (NCLAN) experimental data, tend to support the finding, suggested in the

literature, that the repeated occurrence of hourly average O_3 concentrations of 0.10 ppm and higher result in adverse effects on vegetation. Although the hourly average concentrations below 0.10 ppm may be important in affecting crop yield, the NCLAN program was not developed to identify and quantify the specific exposure regimes that are responsible for the observed effects. In our analysis, we have presented exposure statistics to provide a variety of choices that allow investigators the opportunity to develop indices that are most relevant in predicting vegetation effects.

It has been assumed by some investigators that the O_3 exposures that occurred in the NCLAN chambers during the fumigation period were greater than those received during the remaining part of each day. For example, it has been assumed that the number of hourly average concentrations ≥ 0.06 ppm was much greater during the daylight hours than the late afternoon, evening, and early morning hours. For 22 sets of NCLAN experiments, over the entire exposure period, we have compared the SUM06 value calculated over the daily exposure period (e.g., 7 and 12 hours) with the SUM06 value calculated over a 24-h period. Assuming that the ambient hourly average concentrations reported for each experiment represented the exposure the crops received during those periods when fumigation did not occur, we combined these data with the fumigation-period information reported by the investigators for each chamber.

In most cases, the 24-h SUM06 values for the lower exposure chambers were more influenced by hourly average concentrations ≥ 0.06 ppm that occurred outside the daily fumigation period than the 24-h SUM06 values for the higher O_3 exposure treatments. The value calculated for the SUM06 index over the exposure period did not necessarily represent the 24-h SUM06 value. Thus, if one ignores the hourly average concentrations ≥ 0.06 ppm that occurred outside

the fumigation period, exposure-response equations developed using only fumigation-period air quality data, would at times, appear to overestimate yield reductions. Thus, there is some degree of uncertainty associated with using the SUM06 exposure index and therefore, inferences based on the published exposure-response results should be used with caution.

The problems associated with using long-term seasonal average concentrations, such as 7-h seasonal values, as surrogates for dose are well documented. Any O_3 exposure index used to describe those regimes that cause vegetation effects must be able to characterize adequately the upper tail of the hourly average distribution curve. The cumulative exposure indices, SUM06 (i.e., the sum of all hourly average concentrations ≥ 0.06 ppm) and W126 (i.e., the sum of all hourly average concentrations where the higher concentrations receive greater weight than the lower values), have shown much promise in relating O_3 exposure with vegetation effects.

However, even if one is found to characterize the most important components of exposure (e.g., the upper tail of the hourly average distribution curve), a consistent relationship between an O_3 exposure index and vegetation effects may not always occur. We know, based on published results in the literature, that the occurrences of elevated O_3 hourly concentrations are important for eliciting adverse effects on agricultural crops. However, in addition to concentration, the (1) amount and chemical form of the pollutant that enters the target organism, (2) length of the exposure within each episodic event, (3) time between exposures (i.e., the respite or recovery time), and (4) sensitivity of the target organism are important factors that affect vegetation. When predicting vegetation effects, it is unclear how important these four factors are in an overall weighting

scheme. However, at this time, given the current state of knowledge, concentration should be weighted more heavily than either sensitivity or actual dose.

For protecting vegetation from O_3 exposures, an important aspect that requires further attention is the use of experimental results obtained at low-elevation sites to predict O_3 vegetation effects that may occur at high-elevation locations. Ozone exposures that occur at high-elevation sites are often different from those that occur at lower elevation locations. Exposure regimes used in experiments performed at low-elevation locations should mimic those that occur at the high-elevation sites. In addition, the use of mole fraction (e.g., ppm) or absolute concentration (e.g., micrograms per cubic meter) to describe exposure is an important consideration. Exposure-response relationships developed using results obtained at low-elevation locations may require pressure adjustments when attempting to use air quality data obtained at high-elevation monitoring sites to predict vegetation effects. When concentrations of gases are defined in terms of mole fraction (i.e., units of ppm), the resulting term is invariant to temperature and pressure. However, if exposures measured at low-elevation sites are compared with those experienced at high-elevation sites, the variation of concentration (in units of micrograms per cubic meter) as a function of altitude may be significant. Given the same parts-per-million value experienced at both high- and low-elevation sites, the absolute concentrations (i.e., micrograms per cubic meter) at two elevations are different. Temperature decreases inversely relative to elevation and therefore, the change in absolute concentration would be less than estimated when only pressure changes are considered. However, temperature differences do not usually compensate for the pressure

effect. The biological consequences of high-elevation exposures to the reduced absolute concentration of O_3 that are disguised by the use of mole fraction units of concentration need to be further investigated.

Because of the concern that the current form of the standard may not protect vegetation from O_3 effects, we have explored the effects on nonattainment status by lowering or modifying the current form. When exploring the effects on nonattainment status when the current form of the standard was changed from 0.12 ppm to either 0.10 or 0.08 ppm for the 1987-89 and 1986-88 periods, we found the greatest increase in nonattainment areas occurred when the standard was lowered to 0.10 ppm. The application of a revised standard for O_3 would mainly increase the number of nonattainment areas (i.e., CMSA, MSA, and non-MSA) that are not near the current existing areas. In other words, rather than growth occurring near existing nonattainment areas, it would occur at new locations removed from the current nonattainment areas.

Except for the Plains States, the major growth on a regional basis would be dramatic for all regions across the United States. The most dramatic differences would be in regions where states were completely in attainment with the current standard. For example, Oregon and Washington were in attainment for the 1987-89 period. However, if a standard of 0.10 ppm were applied, the Seattle/Tacoma, Portland, and Eugene areas would be classified as nonattainment. All Rocky Mountain states, other than the Salt Lake area of Utah, are currently in attainment. A revised standard would classify the Denver, Phoenix, and Las Cruces areas into nonattainment status.

As indicated, results reported in the literature indicate that the second highest daily maximum concentration appears to be an inappropriate

index to use to protect vegetation from elevated O_3 exposures. As an alternative to the current form of the standard, it has been suggested that the SUM06 O_3 exposure index could be used as the form of a secondary standard to protect agricultural crops. It has been reported in the literature that a 3-month SUM06 value of 24.4 ppm-h was estimated to cause a 10% yield loss in some NCLAN experiments.

Accordingly, we identified those areas in the United States that experienced a SUM06 value of 24.4 ppm-h or higher over a 3-month period for the years 1987, 1988, and 1989. We explored whether there might exist a relationship between the current form of the standard, lowered to either 0.10 or 0.08 ppm, and the SUM06 3-month cumulative index. Based on our results, lowering the current form of the standard to either 0.10 or 0.08 ppm did not appear to guarantee that a specific monitoring site would achieve a SUM06 3-month cumulative value of 24.4 ppm-h or lower.

In addition, we found that the occurrence of 3-month SUM06 values of 24.4 ppm-h or higher was not correlated with elevated hourly average concentrations and concluded that the application of the SUM06 index as a secondary standard would result in inconsistent protection for vegetation. Using 1989 hourly averaged O_3 data, we found that no strong relationship appeared to exist between the number of occurrences of high hourly average O_3 and a maximum uncorrected 3-month SUM06 value ≥ 24.4 ppm-h. Several O_3 monitoring sites that violated the current standard experienced a 3-month SUM06 value < 24.4 ppm-h. Similarly, we found that several O_3 monitoring sites that did not violate the current standard experienced a maximum uncorrected 3-month SUM06 value ≥ 24.4 ppm-h.

As indicated, we found that a strong correlation between peak concentrations and the value of the SUM06 index did not necessarily occur under ambient conditions. However, as reported in the literature, the SUM06 index has performed well, using NCLAN data, in relating O_3 exposure and yield reduction. We found, at the 20% yield reduction level, that there were O_3 distributions (of hourly average concentrations) which contained a sufficient number of high hourly average concentrations. The NCLAN experimental protocol applied incremental and proportional additions that resulted in many of the treatments experiencing elevated O_3 exposures; many of the artificial regimes used by NCLAN contained the elevated hourly average concentrations that were reflected in the determination of the absolute values of the cumulative indices. Therefore, at many of the treatment levels, the magnitude of the SUM06 index, calculated using NCLAN protocols, appeared to be influenced by the peak exposures that correlated well with the observed growth reductions.

A major concern about the use of any exposure index (e.g., cumulative or seasonal average concentration) is whether the value of the index can be linked to a specific exposure regime. The absolute value of the index reflects only the mathematical calculation performed using hourly average O_3 concentrations. If we assume that the distribution of the highest hourly average concentrations (i.e., the upper tail of the distribution) is an important factor in affecting vegetation, then a single-parameter exposure index, such as the SUM06 or W126, in some instances, may not be specific enough to describe those important distributions that cause an O_3 -related effect.

Although difficulties may exist for linking experimental exposure-response relationships with ambient air for predicting vegetation effects,

single-parameter exposure indices have been used successfully for describing regional O_3 exposure in the United States. Yet, given the fact that we have shown that the magnitude of cumulative exposure indices, such as the W126 or SUM06 exposure index, is not necessarily strongly associated with the occurrence of high hourly average O_3 concentrations, why is it possible to successfully describe regional exposures using single-parameter cumulative indices?

The O_3 exposures experienced at each site are influenced by a multitude of factors. The elevation of a specific site, its ground cover (i.e., sorptive capacity), as well as its latitude, may influence O_3 production and destruction of the absolute O_3 exposure value experienced at a specific site. Many of the O_3 monitors used in the kriging analyses were situated near urban-oriented locations. Thus, the distribution of the hourly average concentrations may have been similar. For example, most of the urban-oriented monitoring sites may experience similar scavenging processes that result in 30% or more of the hourly average concentrations occurring below 0.015 ppm. In addition, the maximum hourly average concentrations experienced at many of these sites were similar. Thus, with similar hourly average distribution patterns, it would be assumed that the magnitude of a cumulative exposure index, such as the W126 or SUM06, would order itself properly, with the higher value corresponding to the higher exposure. This appears to be what occurred.

In addition to using cumulative exposure indices to describe regional O_3 exposures, a cumulative exposure index has been used in trends analysis. Trends for O_3 exposures over 5- and 10-year periods (i.e., 1984-1988 and 1979-1988) have been summarized for rural locations in the United States. The evidence for trends at each monitoring location was explored. Evidence for

regional trends was based on studying the individual time trends observed for each of the sites in the region. The seasonal W126 cumulative exposure index was used to investigate trends. The results reported in the literature were consistent with the findings reported by the U.S. Environmental Protection Agency.

The explanation for the successful application of the cumulative index in the trends analysis was similar to the one given for the kriging analysis. For a specific monitoring site, the hourly average distribution pattern was similar over the years studied. The scavenging processes remained the same over time at a specific site. Thus, the difference in magnitude of the W126 index, at any one site over time, was reflected in changes in the distribution curve of the hourly average O_3 concentrations. Changes that occurred at the upper end of the distribution curve were reflected in the magnitude of the W126 index.

For some purposes, the single-parameter index appears to work appropriately. However, the predictive power involving exposure-response relationships that use single-parameter exposure indices may not be as strong as desired. A multiple-parameter index may be necessary to adequately describe distribution patterns of hourly average concentrations. To improve the predictive capability that depends upon linking experimental exposure-response relationships with ambient air quality, it appears that indices, such as the SUM06 or W126, will have to be combined with other exposure parameters in order to mathematically define unique distribution patterns of hourly average concentrations.

Although moderate success has been achieved using the SUM06 and W126 exposure indices, consistency is important so that experimental exposure-

response relationships can be strongly linked with ambient exposures. If this consistency is not present, then it will be difficult to use any exposure index in the development of a secondary standard.

For developing a secondary standard to protect vegetation, the combined exposure statistics should be selected based on the observation that high concentrations are expected to cause greater impact on vegetation than lower concentrations. It has been shown, when high hourly average concentrations are present in an exposure regime, that single-parameter cumulative indices can be used to relate O_3 exposures with vegetation growth reductions. However, when attempting to link experimental models with ambient air quality, it appears that the application of a single-parameter exposure index, in the form of a standard for protecting vegetation, will provide inconsistent results. This does not imply that all currently used cumulative exposure indices are not appropriate for describing O_3 exposure. Rather, it appears that cumulative indices, such as the SUM06 and W126 indices, will have to be combined with other parameters to quantify accurately the occurrence of the high hourly average concentrations.

The possible combination of exposure parameters, such as the (1) sigmoidally-weighted exposure index or (2) SUM06 index, with other indices should provide sufficient means to describe those unique distribution curves that have the potential for eliciting an adverse effect. Our reanalysis of the NCLAN data provided us with evidence that summaries of distribution patterns provide important information concerning the relationships between exposure and response. Future research efforts in this area point to the quantification of the distribution of the hourly average concentrations. The percentile distribution of the hourly average concentrations offers a way to

characterize both high and low O_3 concentrations. With high confidence, from the percentile distribution of O_3 , one can infer that the values in the tail of the distribution represent peaks in the time plots of hourly O_3 concentrations.

In addition, percentile distributions offer the opportunity to differentiate exposures experienced at remote or isolated sites from exposures experienced at sites influenced by urban sources. Monitoring sites under the influence of local urban sources experience approximately 50-70 percent of their hourly average O_3 concentrations above 0.015 ppm.

Although we have discussed the possible combinations of parameters to better link experimental exposure-response models with ambient air quality for predicting possible impacts on vegetation, at this time, information is not available to identify the specific parameters that should be combined. However, the results of the NCLAN experiments provide researchers with the opportunity to better understand the level of exposures that result in agricultural yield reduction. We have summarized the distribution of the hourly average concentrations that occurred in some of the NCLAN experiments. The characterized distributions reflected the importance of the upper end of the distribution curve in affecting crop yield reductions. We believe this additional information should assist researchers in identifying a multi-parameter exposure index that will properly relate ambient exposure to response.

A strong case has been made for selecting multi-parameter exposure indices for establishing a secondary standard to protect vegetation from high levels of O_3 exposure. However, caution is urged. Although we believe that an effort should be made to identify multi-parameter indices, it is important

to note that a consistent relationship between multi-parameter exposure indices and vegetation effects may not always exist. Based on the analysis described in this report, at this time, we believe that further research is required before any single-parameter exposure index is used in the standard-setting process to protect vegetation from O_3 exposure.

CHAPTER 1

INTRODUCTION

1.1 BACKGROUND

The Clean Air Act requires the Administrator of the U.S. Environmental Protection Agency to establish national ambient air quality standards. These standards are designed to protect the public health and welfare from any known or anticipated adverse effects associated with the presence of criteria air pollutants. Primary air quality standards are promulgated to prevent adverse effects on human health, while secondary air quality standards are established to prevent adverse welfare effects (e.g., effects on vegetation, animals, deterioration of property materials, and visibility).

The ubiquity and toxicity of ambient air O_3 is well documented (EPA, 1986, 1988a). Because O_3 is an omnipresent air pollutant that affects both human health and vegetation, the U.S. Environmental Protection Agency (EPA) has established both primary and secondary standards.

On April 30, 1971, in the Federal Register (36 FR 8186), the Environmental Protection Agency promulgated National Ambient Air Quality Standards (NAAQS) for photochemical oxidants. The scientific, technical, and medical bases for these standards were contained in the air quality criteria documents for photochemical oxidants, published by the U.S. Department of Health, Education, and Welfare in March 1970. Both the primary and secondary standards were set at an hourly average level of 0.08 ppm, not to be exceeded more than once per year.

Based on a reassessment of the available data, in 1979, EPA revised both the primary and secondary standards for photochemical oxidants (i.e., O_3). The revised form of the standard (1) raised the primary standard to 0.12 ppm,

(2) raised the secondary standard to 0.12 ppm, and (3) changed the definition of the point at which the standard is attained to "when the expected number of days per calendar year with maximum hourly average concentrations above 0.12 ppm is equal to or less than one." The phrase "expected number of days per calendar year" differed from the previous NAAQS for photochemical oxidants, which simply stated a particular concentration "not to be exceeded more than once per year." The federal standard for O_3 is based on the second daily occurrence of a maximum hourly average concentration above 0.12 ppm and is designed to protect both human health and welfare effects.

There is no requirement that the primary and secondary standards be identical, nor is there any requirement that only a single expression of the standard be used (i.e., an average concentration for a single time period versus multiple exceedances or integrated exposures). Any effort to propose a secondary standard, whose form is different than the current form of the primary and secondary standard, implies that either (1) the current form is inappropriate for protecting the public welfare or (2) a more restrictive value of the current form of the standard is required.

There have been indications reported in the literature (Lefohn *et al.*, 1989; Lee *et al.*, 1991) that the current form of the standard may not be appropriate for protecting vegetation from O_3 exposures. Lee *et al.* (1991) reported that, although no single exposure index was best in describing the exposure-response relationship for 49 case studies, the performance of the current form of the U.S. Federal standard was considerably worse than other exposure indices used in their analysis. The authors reported that the current form of the standard did not perform adequately because it (1) was

poorly related to plant growth, (2) ignored exposure duration, and (3) placed too much emphasis on a single peak 1-h concentration.

Should one want to develop an O_3 standard that provides an adequate measure of protection to vegetation, it would be necessary to define, in as precise terms as possible, the relationship between O_3 exposures and the potential for adverse effects on vegetation. Although the form of the standard should be made as simple as possible, it is essential that the standard be related directly or indirectly to identifiable adverse effects. The U.S. EPA (1988b) has made a distinction between the relative importance of foliar injury to vegetation and reduced crop yield. Greater emphasis has been placed on damage or yield loss than on injury, where injury encompasses all measurable plant reactions, such as reversible changes in metabolism, reduced photosynthesis, leaf necrosis, leaf drop, altered quality, or reduced growth, that do not influence agronomic yield or reproduction and damage includes all effects that reduce the intended human use or value of the plant or ecosystem (Tingey *et al.*, 1990).

The purpose of this report is to identify and review some of the key issues related to assessing the effects of O_3 on vegetation. Our report has reviewed the available information on (1) components of O_3 exposure that elicit adverse effects on vegetation, (2) ways to describe these components in the form of O_3 exposure indices that may be useful in the standard-setting process for protecting vegetation, (3) the change in nonattainment status that may occur should the existing O_3 standard be modified, and (4) the need for future research efforts to explore the development of a multi-parameter index to protect vegetation from O_3 exposure.

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CHAPTER 2

EXPOSURES THAT RESULT IN VEGETATION GROWTH REDUCTION

2.1 INTRODUCTION

For assessing the efficacy of a standard to protect vegetation from O_3 , the actual pollutant levels below which plants will be protected must be identified. Guderian *et al.* (1985) have pointed out that during chronic exposures, injury increases with increasing concentration and that plant growth is influenced more by concentration than exposure duration, when similar products of concentration and time are used. Similar results have been published relating O_3 exposure to vegetation growth reduction.

The importance of high hourly average O_3 concentrations affecting vegetation growth has been documented (U.S. EPA, 1986). Short-term, high concentrations have been identified as being more important than long-term, low concentrations (Heck *et al.*, 1966; Heck and Tingey, 1971; Bicak, 1978; Henderson and Reinert, 1979; Nouchi and Aoki, 1979; Reinert and Nelson, 1979; Bennett, 1979; Stan *et al.*, 1981; Musselman *et al.*, 1983, 1986; Ashmore, 1984; Amiro *et al.*, 1984; Tonneijck, 1984; Hogsett *et al.*, 1985a). Similarly, for trees, high concentrations appear to be an important factor (Hayes and Skelly, 1977; Mann *et al.*, 1980; Hogsett *et al.*, 1985b).

Although all plants are capable of being adversely affected by exposure to phytotoxic gases and particulates in polluted air, the nature of the response can be extremely variable. Runeckles and Wright (1988) have indicated the following features play important roles in determining target sensitivity:

- the species of plant;
- the stage of development of the plant;

- the nature of the pollutant or mix of pollutants;
- the pattern of exposure to the pollutant(s), which involves consideration of the concentration and durations of exposure;
- environmental conditions in the soil, such as water availability and nutrition;
- environmental conditions in the ambient air, such as light intensity, temperature, humidity, and air movement; and
- biological factors, such as the occurrence of pests and diseases, and competitive stresses exerted by individual plants on their neighbors.

For estimating levels that are required to protect vegetation from O_3 exposures, it is necessary to take into consideration the large variability in response. This chapter discusses the ranges of O_3 exposures that result in injury and damage to vegetation, as well as exposure indices that warrant further consideration as possible surrogates for dose in the standard-setting process.

2.2 OZONE EXPOSURES THAT AFFECT YIELD REDUCTION

Guderian *et al.* (1985) have proposed maximum acceptable O_3 concentrations for the protection of vegetation. The authors' numerical values are based on the limiting values proposed by Jacobson (1977) and the exposure-response values for definite injury levels developed by Heck and Brandt (1977). In general, the recommendations made by Guderian *et al.* (1985) appear to reinforce the belief that hourly average concentrations of 0.10 ppm and higher are required to elicit adverse effects on vegetation.

The one exception to the recommendations made by Guderian *et al.* (1985) was for the protection of sensitive species. The authors recommended that sensitive vegetation should not be exposed for more than 4 hours to hourly average concentrations of 0.05 ppm. Ozone hourly average concentrations of

0.05 ppm routinely occur at many "clean" site locations in the world (Lefohn *et al.*, 1990a). The occurrence of hourly average concentrations of 0.05 ppm are not necessarily associated with anthropogenic sources and thus, using a threshold of 0.05 ppm may not be realistic for protecting sensitive species. Table 2-1 summarizes the recommendations made by the authors for hourly average concentrations for durations of exposures of 0.5, 1, 2, and 4 hours. The information in the table provides an indication that long-term exposures consisting only of lower hourly average O_3 concentrations will not necessarily produce adverse effects on vegetation.

The National Crop Loss Assessment Network (NCLAN) program represents one of the most extensive data bases in existence for identifying O_3 exposure regimes that may elicit an adverse effect on crops. NCLAN was initiated and sponsored by the U.S. Environmental Protection Agency to evaluate the effects of O_3 on the productivity of major regional crops under field conditions. Open-top chambers were used to introduce artificial O_3 exposures. For the period 1980 through 1986, NCLAN investigators exposed several different crops to O_3 exposures to identify levels at which crop reduction occurred. Table 2-2 summarizes the different crops and periods of exposure.

The limitations of the NCLAN methodologies have been described elsewhere (Lefohn and Runeckles, 1987; Krupa and Kickert, 1987; Lefohn *et al.*, 1988; Lee *et al.*, 1988; Heuss, 1982; Krupa, 1985; Brennan *et al.*, 1987; Smith *et al.*, 1987; Ashmore, 1988; Runeckles and Wright, 1988). Some of the more important limitations summarized by Lefohn *et al.* (1989) are

- Even though high ambient hourly O_3 concentrations are observed during 1200-2000h at agricultural sites in much of the U.S. during the crop growth season, the NCLAN experiments were designed with exposures to added O_3 in the open-top chambers between 0900-1559h or, in the final years of the program, 0900-2059h. When the 0900-1559h, 7-h period was used,

frequent high O_3 ambient exposures beyond 1559h were excluded from calculations of exposure indices, although the crops received these ambient exposures;

- The relative differences between the O_3 exposure treatments were always constant. In most cases, O_3 was added to the chambers when the hourly average concentration exceeded 0.03 ppm. Thus, the plants in the higher O_3 treatments were given little opportunity to recover from stress. Under actual ambient conditions, O_3 concentrations vary in time and space and periods occur when exposures are both high and low (Runeckles and Wright, 1988);
- In the exposure treatments with highest O_3 , in the cases examined, the frequency distribution of hourly O_3 within the chambers showed a bimodal distribution (Lefohn *et al.*, 1988) or even a polymodal distribution (Heagle *et al.*, 1986). Ambient O_3 follows a unimodal distribution;
- In some cases, infrequent sampling of O_3 within a given hour has resulted in uncertainty and controversy regarding the accuracy of the published hourly average O_3 values;
- In analyzing NCLAN data and establishing cause-and-effect relationships, a number of exposure parameters and models were tested (refer to Heck *et al.*, 1988). In the end, the Weibull function was selected as providing the most suitable empirical exposure-response model. Since experimental results were obtained first and the model fitted afterwards, concern may be raised as to whether the best-fit model is a product of the specific NCLAN experimental design. The Weibull model performed differently at different NCLAN sites. Furthermore, it was unable to explain one set of independent results (Brennan *et al.*, 1987; Smith *et al.*, 1987);

The 7-h (0900-1559h) average, calculated over an experimental period, was used to summarize O_3 exposures by the NCLAN program (Heck *et al.*, 1982). The 7-h daily daylight period was selected by NCLAN because the parameter was believed to correspond to the period of greatest plant susceptibility to O_3 pollution. In addition, the 7-h period of each day (0900-1559h) was assumed to correspond to the time that the highest hourly O_3 concentrations would occur. In later years, the 12-h average, calculated over an experimental period, was used to describe O_3 exposures. In the published literature, the

majority of NCLAN's experiments were summarized using the 7-h experimental-period average and other long-term average concentration statistics.

In retrospective studies using NCLAN data, attempts were made to explore the efficacy of alternative O_3 exposure statistics in describing the relationship between exposure and response (Lefohn *et al.*, 1988; Lee *et al.*, 1988, 1989, 1991). Because the retrospective studies mainly focused on the adequacy of mathematical parameters to relate exposure with growth reduction, no attempt was made to describe the specific O_3 exposure regimes that elicited an adverse effect on vegetation. It was assumed that the mathematical parameters adequately correlated with the important components of exposure that elicit an adverse effect. As will be discussed in a later chapter, the absolute value associated with an exposure index does not necessarily correlate with the important components of exposure. Therefore, we investigated the O_3 exposures that occurred in the NCLAN experiments for which a specific level of growth reduction was observed.

Lee *et al.* (1991), using vegetation effects data obtained from 31 field experiments (involving 12 crops), mostly operated by the NCLAN program, evaluated the efficacy of four O_3 exposure indices. Based on a review of the efficacy of the four O_3 exposure indices evaluated by Lee *et al.* (1991), Tingey *et al.* (1991) recommended that the SUM06 O_3 exposure index could be applied as the form of a secondary standard to protect agricultural crops. The authors reported that a 3-month SUM06 value of 24.4 ppm-h was estimated to cause a 10% yield loss in half the cases they investigated.

As a part of their analysis, the investigators developed, using the SUM06 index (the sum of all hourly average concentrations ≥ 0.06 ppm over the exposure period), exposure-response models that predicted yield reduction. In

most cases, Lee *et al.* (1991) used only the artificial fumigation period (e.g., 7- and 12-h periods) to determine the SUM06 value. The investigators assumed that the period outside the fumigation window (i.e., the 17 and 12 hours, respectively) did not contribute greatly to the SUM06 value. Based on the Lee *et al.* (1991) equations, Table 2-3 summarizes the predicted yield loss, using the SUM06 value of 24.4 ppm-h.

Lee *et al.* (1991) assumed a SUM06 value of 0.00 ppm-h at 100% yield. We explored the validity of using a 3-month cumulative SUM06 value of 0.00 ppm-h. Lefohn and Foley (1991) have characterized O₃ hourly average concentration data collected at several national park locations and have compared these data with several "clean" O₃ monitoring sites (Lefohn *et al.*, 1990a). Using hourly average O₃ data from six national park sites (Glacier, Great Sand Dunes, Yellowstone, Badlands, Theodore Roosevelt, and Arches) and two national forest locations (Custer and Ochoco), the SUM06 3-month cumulative value was determined over a 24-h window period (Table 2-4). The average 3-month cumulative SUM06 value over the 16 site-years was 3.07 ppm-h. This value was used in the equations developed by Lee *et al.* (1991) and the results compared with the predicted yield loss that resulted when an assumed SUM06 value of 0.00 ppm-h was used at the 100 yield point. As indicated in Table 2-3, the "correction factor" is small and therefore, an assumed SUM06 value of 0.00 ppm-h for "clean" site locations does not result in large discrepancies when compared with the predicted yield losses when a SUM06 value of 3.07 ppm-h is used.

As indicated above, Lee *et al.* (1991) assumed that the SUM06 value was not greatly influenced by the O₃ exposures that occurred outside the 7- and 12-h daylight period when fumigation occurred. The investigators assumed that

the number of hourly average concentrations ≥ 0.06 ppm was much greater during the daylight hours than the late afternoon, evening, and early morning hours. For 22 sets of NCLAN experiments, over the entire exposure period, we have compared the SUM06 value calculated over the daily exposure period (e.g., 7 and 12 hours) with the SUM06 value calculated over a 24-h period. Assuming that the ambient hourly average concentrations reported for each experiment represented the exposure the crops received during those periods when fumigation did not occur, we combined these data with the fumigation-period information reported by the investigators for each chamber.

As anticipated, in most cases, the 24-h SUM06 values for the lower-exposure chambers were more influenced by hourly average concentrations ≥ 0.06 ppm that occurred outside the daily fumigation period than the 24-h SUM06 values for the higher O_3 exposure treatments (Table 2-5). The value calculated for the SUM06 index over the exposure period did not necessarily represent the 24-h SUM06 value. Thus, if one ignores the hourly average concentrations ≥ 0.06 ppm that occurred outside the fumigation period, the exposure-response equations developed by Lee *et al.* (1991), at times, appear to overestimate yield reductions. Because, in most cases, the form of the model used by Lee *et al.* (1991) is dependent on several variables, it is unclear if the overestimation would affect the entire range of O_3 exposures or only the lower exposures.

We have summarized the O_3 exposures, by treatment level, that occurred in 22 NCLAN experiments (Table 2-6). Because the exposures within each chamber, at a specific treatment, were similar within an experiment, we have presented one chamber per treatment per experiment in order to summarize the exposure statistics. No attempt was made to combine similar treatments within

an experiment and pool the results. The exposure statistics were determined over a 24-h time frame during the exposure period.

Although there were inaccuracies associated with using the exposure-response models developed by Lee *et al.* (1991), we applied the models to obtain rough estimates of yield reduction. The exposure-response models were used to calculate SUM06 cumulative exposures that produced 10%, 20%, and 30% yield reductions for a subset of the NCLAN experiments (Table 2-7). The values of the SUM06 cumulative exposures that produced a specific yield reduction (i.e., 10%, 20% and 30%) were compared with the treatment levels that occurred within each experiment to identify those exposure regimes that may have been responsible for the crop reduction (see Tables 2-6 and 2-7). Because of the uncertainty associated with the yield predictions, we summarized the exposure statistics for those treatments that predicted approximately 20% yield reduction (Table 2-8), recognizing that the yield reduction would more than likely be less than the 20% predicted. In most cases, the SUM06 value listed in Table 2-7 in the 20% reduction column could not be matched with the SUM06 value experienced in a specific treatment. Therefore, the summary statistics from the treatment that experienced the SUM06 value closest to the value listed in Table 2-7 were used in Table 2-8. Most of the identified exposure regimes were associated with treatments where O_3 had been incrementally or proportionally added into the chamber. In approximately 85% of the cases, the SUM06 cumulative exposure value used, which was determined over the fumigation period, represented more than 85% of the actual value experienced over the 24-h period.

In general, repeated exposures of hourly average concentrations ≥ 0.10 ppm occurred in most of the treatments identified in Table 2-8. Similar to

the results reported by Lee *et al.* (1991), soybean data predominated the analysis. Reviewing the results for soybean, we found, in most cases, that at least 5% of the hourly concentrations in the identified exposure regimes were ≥ 0.10 ppm in the NCLAN open-top chambers. The frequency of occurrence ≥ 0.10 ppm ranged from 5 to 517 and the maximum hourly average concentrations in the experiments ranged from 0.123 ppm to 0.292 ppm.

For wheat, an inconsistent result occurred. Because Vona wheat is extremely sensitive to O_3 exposures (EPA, 1986), ambient O_3 exposures were predicted to cause a 20% yield reduction. However, as noted in Table 2-5, 54% and 40% of the SUM06 values experienced in the NF treatments in 1982 and 1983, respectively, occurred outside the 7-h exposure period window. Thus, the application of the SUM06 model determined by Lee *et al.* (1991) would result in an overestimate of yield reduction. For Abe and Arthur, we found that NCLAN experimental exposures with large numbers of hourly average concentrations ≥ 0.10 ppm (i.e., 186) resulted in a predicted 20% yield reduction.

Tobacco and peanut appeared to be more sensitive to O_3 exposure than cotton (Table 2-8). In addition, corn and sorghum appeared to be highly resistant to O_3 exposure.

Our results, using a select set of NCLAN experimental data, tend to support the finding suggested by Guderian *et al.* (1985) that the repeated occurrence of hourly average O_3 concentrations of 0.10 ppm and higher result in adverse effects on vegetation. In our analysis, we subjectively used a 20% yield reduction threshold. We believe that using a lower yield reduction threshold would not be appropriate at this time because of all the uncertainties mentioned previously. Although the hourly average concentrations below 0.10 ppm may have been important in affecting crop yield

in the experiments, the NCLAN program was not developed to identify and quantify the specific exposure regimes that are responsible for the observed effects. Thus, at this time, we believe that the approach we have used makes it possible for those who are interested to establish secondary standards that will protect vegetation from O_3 exposures. The exposure statistics presented in Table 2-8 provide a variety of choices that allow investigators the opportunity to develop indices that are most relevant in predicting vegetation effects.

2.3 SELECTING APPROPRIATE EXPOSURE INDICES

In the previous section, the discussion focused on the components of O_3 exposure that elicit adverse effects on vegetation. In this section, the focus turns toward ways to accurately describe these components in the form of exposure indices that may be useful in the standard-setting process for protecting vegetation.

As discussed in the previous section, there are a number of ways in which hourly O_3 concentrations can be summarized. The selection of suitable measures for defining the "dose" term in exposure/dose-response relationships is an important aspect that has received considerable discussion (U.S. EPA, 1986; Hogsett *et al.*, 1988; Lefohn *et al.*, 1989; Lefohn *et al.*, 1990b). Any index that is selected as a surrogate for "dose" should (1) describe the most important exposure characteristics that elicit an adverse effect and (2) order itself properly when comparing the absolute value experienced in an experiment, with the value calculated under actual ambient conditions.

Exposure indices are important because they form the linkage between air quality standards that are promulgated to protect specific targets and the

actual dose that is responsible for eliciting an effect. Results have been reported in the literature relating O_3 exposure with vegetation effects. Although the perfect exposure index that can serve as a surrogate for dose does not exist, there are some O_3 exposure indices that do relate fairly well with vegetation effects (Lefohn *et al.*, 1988; Lefohn *et al.*, 1990b; Lee *et al.*, 1988, 1989, 1991).

For almost seventy years, air pollution specialists have explored alternative mathematical approaches for summarizing ambient air quality information in a form that can serve as a surrogate for dose. For assessing the possible effects of O_3 on agricultural crop and forest, researchers have focused on characterizing 1-h average values in "biologically meaningful" forms. Obtaining a definition of "biologically meaningful" from several different effects researchers is a difficult task. However, based on biological evidence, it is clear that any parameter used as a dose surrogate for predicting vegetation effects should focus on the upper tail (i.e., the highest hourly average concentrations) of the distribution curve.

For vegetation, there has been considerable effort to identify ways to describe O_3 exposures that elicit adverse effects (EPA, 1986; Lefohn and Runeckles, 1987; Krupa and Kickert, 1987; Hogsett *et al.*, 1988; EPA, 1988a; Lefohn *et al.*, 1989; Lefohn *et al.*, 1990b). Since the early 1980s, there has been much discussion concerning the importance of the higher hourly average concentrations in relationship to the lower concentrations (EPA, 1986; Lefohn and Runeckles, 1987; Lefohn *et al.*, 1989; Lefohn *et al.*, 1990b). Several different types of exposure indices have been proposed.

A 6-h long-term seasonal average O_3 exposure parameter was used by Heagle *et al.* (1974). Also, Heagle *et al.* (1979) reported the use of a 7-h

experimental-period average. As indicated earlier in this chapter, the 7-h (0900-1559h) average, calculated over an experimental period, was adopted as the statistic of choice by the U.S. EPA's National Crop Loss Assessment Network (NCLAN) program (Heck *et al.*, 1982). Toward the end of the program, NCLAN redesigned its experimental protocol and applied proportional additions of O_3 to its crops for 12-h periods. The expanded 12-h window reflected NCLAN's desire to capture more of the daily O_3 exposure.

In the 1980s, concerns about the use of a long-term average to summarize exposures of O_3 appeared in the literature (Lefohn and Benedict, 1982; Tingey, 1984; Lefohn, 1984; Lefohn and Tingey, 1985). Long-term seasonal average concentrations (e.g., 7-or 12-h average concentrations) did not correlate strongly at most O_3 monitoring sites with the components of exposure regimes that were most important in affecting vegetation. EPA (1986) noted that the weight of evidence appeared to suggest that long-term averages, such as the 7-h seasonal average, were not adequate indicators for relating O_3 exposure and plant response. EPA (1988b) pointed out that repeated peak concentrations appeared to be the most critical element in determining plant response, and the Agency indicated that exposure indicators which emphasize peak concentrations and accumulate concentrations over time, probably provide the best biological basis for standard setting.

Searching for an alternative to the long-term average concentration parameter, Lefohn and Benedict (1982) introduced an exposure parameter based on the hypothesis that if the higher O_3 concentrations were more important in eliciting adverse effects on agricultural crops than the lower values, then the higher hourly mean concentrations should be given more weight than the lower values. This integrated exposure parameter summed all hourly

concentrations equal to and above a threshold level (i.e., 0.10 ppm). The exposure parameter was similar to that used by Oshima (1975), where the difference between the value above 0.10 ppm and 0.10 was summed.

In the late 1980s, the focus turned from the use of long-term seasonal averages to cumulative indices (e.g., exposure parameters that sum the products of concentrations multiplied by time over an exposure period). Besides the cumulative indices proposed by Oshima *et al.* (1976) and Lefohn and Benedict (1982), other cumulative indices, such as (1) the number of occurrences of daily maximum hourly averaged concentrations greater than a threshold level (Ashmore, 1984) and (2) the use of exponential functions (Nouchi and Aoki, 1979; Larsen and Heck, 1984) to assign unequal weighting to O_3 concentrations were suggested.

The use of the integrated exposure index, as defined by Oshima (1975) and Lefohn and Benedict (1982), had limitations. The parameter ignored the lower hourly mean concentrations. Early evidence for testing cumulative indices came from results reported by Oshima *et al.* (1976). Similarly, Lefohn and Benedict (1982), applying their cumulative integrated exposure index, reported fairly good agreement between exposures of O_3 and predicted agricultural yield loss in California. The two exposure indices apparently performed well because of the frequent occurrence of high hourly mean O_3 concentrations (e.g., ≥ 0.10 ppm) and possibly, the short period between episodes. The high frequency of such concentrations was responsible for the magnitude of the cumulative index, as well as the impacts on agricultural crops, and thus, a favorable correlation existed between the index and the agricultural effect.

NCLAN data offered the opportunity to test the hypothesis that cumulative indices that describe O_3 exposures may adequately serve as a dose surrogate for describing exposure/dose-response relationships for agricultural crops. Retrospective studies were performed using NCLAN data (Lefohn *et al.*, 1988; Lee *et al.*, 1988, 1989, 1991).

Lefohn *et al.* (1988), using wheat and soybean data sets summarized by Kohut *et al.* (1986, 1987), compared the use of several exposure indices in describing the relationship between O_3 and reduction in agricultural crop yield. Two of the indices used by Lefohn *et al.* (1988) were determined using a sigmoidally-weighted function, as proposed by Lefohn and Runeckles (1987). The sigmoidally-weighted function focused on the higher hourly average concentrations, while retaining the lower and less biologically-effective concentrations. The sigmoidal weighting function was of the form:

$$w_i = 1/[1+M \times \exp (-A \times c_i)]$$

where:

M and A are arbitrary positive constants

w_i = weighting factor for concentration i

c_i = concentration i (in ppm)

The arbitrary positive constants M and A were 4403 and 126 ppm⁻¹, respectively. Their values were subjectively determined to develop a weighting function that (1) focused on hourly average concentrations as low as 0.04 ppm, (2) had an inflection point near 0.065 ppm, and (3) had an equal weighting of 1 for hourly average concentrations at approximately 0.10 ppm and above.

Unlike the seasonal average index, the cumulative indices performed well when data were combined over a two-year period. Lefohn *et al.* (1988) reported that while none of the exposure indices consistently provided a best fit with

the models tested, their analysis indicated that exposure indices that weight peak concentrations of O_3 differently than lower concentrations of an exposure regime could be used in the development of exposure-response functions.

In a more extensive analysis of NCLAN data, Lee *et al.* (1988) fitted more than 600 exposure indices to response data from seven crop studies. For most of the NCLAN experiments used in their analyses, they characterized the daily hourly mean O_3 concentrations that were recorded over the 7-h period (0900-1559h) by the original experimenters. The alfalfa experiments described by Hogsett *et al.* (1985a) collected exposure data over a 24-h period and these data were included in the analysis of Lee *et al.* (1988). Using mostly the 7-h windowed data provided by the NCLAN investigators, the "best" exposure indices were those that applied a general phenologically weighted, cumulative-impact (GPWCI) index with a sigmoid weighting on concentration and a gamma weighting function as a surrogate for changes in plant sensitivity over time. Cumulative indices with various threshold values performed as well as the GPWCIs. Lee *et al.* (1988) reported that mean indices (e.g., 7-h exposure-period means) did not perform well. The authors concluded that the top-performing indices were those whose form (1) accumulated the hourly O_3 concentrations over time, (2) used a sigmoid weighting scheme, which emphasized concentrations of 0.06 ppm and higher, and (3) phenologically weighted the exposure. The authors suggested that lower concentrations should be included, but given lesser weight, in the calculation of the exposure index. In a subsequent analysis using NCLAN data, Lee *et al.* (1989) reported that the phenologically weighted cumulative impact indices, as well as the sigmoidally-weighted integrated index, centered at 0.062 ppm, and the cumulative censored indices that integrated hourly average concentrations of

0.06 and 0.07 ppm or higher, performed at near optimal levels. The results reported by Lefohn *et al.* (1988) and Lee *et al.* (1988, 1989) demonstrated that some cumulative indices could be used in relating O_3 exposure to vegetation effects.

Research results reported by the U.S. EPA and other investigators have illustrated that cumulative exposure indices appear to provide more promise than long-term average concentration exposure indices in relating exposures with vegetation effects (U.S. EPA, 1988b; Lefohn *et al.*, 1990b). Although cumulative indices offer the advantage of focusing on the higher hourly average concentrations, not all cumulative indices achieve this goal. For example, Lefohn *et al.* (1989) pointed out that the cumulative exposure index that sums all hourly average concentrations (SUM0) weights the lower concentrations more than the higher ones. As indicated above, biological results reported in the literature indicate that an appropriate exposure index should emphasize the higher hourly average concentrations.

In Section 2.2, we found that the NCLAN results support the observation that the occurrence of high hourly average concentrations results in measurable yield reduction. In Section 2.3, we found that the use of long-term average concentrations as dose surrogates does not provide sufficient focus on the high hourly average concentrations and that cumulative exposure indices appear to perform well in the development of exposure-response relationships. Based on evidence published in the literature, as well as special analytical studies sponsored by EPA (1988a, b), the use of cumulative indices to describe exposures of O_3 for predicting agricultural crop effects appears to be a more rational approach than the use of long-term seasonal averages.

Although the appeal of a single number, such as a single exposure index, to describe ambient O_3 pollutant exposure is undeniable, there are problems associated with condensing data to such a point that the identification of the important components of exposure are eliminated. The problems associated with using long-term seasonal average concentrations as surrogates for dose were mentioned above. In addition, it is important to point out that a consistent relationship between an O_3 exposure index and vegetation effects may not occur, even if one is found to characterize the most important components of exposure (e.g., the upper tail of the hourly average distribution curve). We know, based on published results in the literature, that the occurrences of elevated O_3 hourly concentrations are important for eliciting adverse effects on agricultural crops. However, in addition to concentration, the (1) amount and chemical form of the pollutant that enters the target organism, (2) length of the exposure within each episodic event, (3) time between exposures (i.e., the respite or recovery time), and (4) sensitivity of the target organism are important factors that affect vegetation. When predicting vegetation effects, it is unclear how important these four factors are in an overall weighting scheme. If both sensitivity and the actual dose that enters the organism are as important as ambient concentration in the weighting scheme, then a given pollutant exposure will elicit varying biological responses at different times for the same crop, as conjectured by Krupa and Teng (1982). However, at this time, given the current state of knowledge, concentration should be weighted more heavily than either sensitivity or actual dose.

2.4 LINKING EXPERIMENTAL RESULTS WITH HIGH-ELEVATION O₃ EXPOSURES

For protecting vegetation from O₃ exposures, an important aspect that requires further attention is the use of experimental results obtained at low-elevation sites to predict O₃ vegetation effects that may occur at high-elevation locations. It is important to note that O₃ exposures at high-elevation sites are often different from those that occur at lower elevation locations. At some high-elevation sites, the highest O₃ exposures occur in the late evening or early morning hours (Berry, 1964; Lefohn and Mohnen, 1986), which produce a diurnal pattern that is distinctly different from that observed at lower elevation sites (Berry, 1964; Stasiuk and Coffey, 1974; Mohnen *et al.*, 1977; Miller *et al.*, 1986; Lefohn and Jones, 1986; Lefohn and Mohnen, 1986). A flat diurnal pattern, which is observed at some high-elevation sites, is usually interpreted as indicating a lack of efficient scavenging of O₃ and/or a lack of photochemical precursors.

It is important to pay specific attention to the types of exposure indices used to describe high-elevation O₃ exposures. Many times the high-elevation diurnal patterns are different from those at the lower elevation sites, because the lowest O₃ hourly average concentrations at many high-elevation sites are near 0.04 ppm (Lefohn and Jones, 1986). The calculated value determined for exposure indices that focus on the lower hourly average concentrations (e.g., SUM0 index) tends to overstate the potential effects of O₃ on vegetation (Lefohn *et al.*, 1992). The absolute value of the SUM0 exposure index is influenced by the large number of values in the mid-concentration range. The mid-concentration range appears not to be as biologically significant as the infrequent occurrence of the higher concentrations.

In addition to the selection of specific exposure indices that are useful for describing O_3 exposures that occur at high-elevation locations, the use of mole fraction (e.g., ppm) or absolute concentration (e.g., micrograms per cubic meter) to describe exposure is an important issue. Lefohn *et al.* (1990c) have pointed out that exposure-response relationships developed using results obtained at low-elevation locations may require pressure adjustments when attempting to use air quality data obtained at high-elevation monitoring sites to predict vegetation effects. When concentrations of gases are defined in terms of mole fraction (i.e., units of ppm), the resulting term is invariant to temperature and pressure. However, if exposures measured at low-elevation sites are compared with those experienced at high-elevation sites, the variation of concentration (in units of micrograms per cubic meter) as a function of altitude may be significant. Given the same parts-per-million value experienced at both high- and low-elevation sites, the absolute concentrations (i.e., micrograms per cubic meter) at two elevations are different. Temperature decreases inversely relative to elevation and therefore, the change in absolute concentration would be less than estimated when only pressure changes are considered. However, temperature differences do not usually compensate for the pressure effect (Lefohn *et al.*, 1990c).

In considering the effect of pressure changes on concentration, EPA (1978) indicated that moles of gaseous pollutant per liter of air was the most useful parameter when considering health effects caused by exposure to air pollution. The same should be true when considering effects of air pollution on vegetation. There are numerous environmental factors that affect the relationship between exposure and response. For example, the amount of O_3 entering the stomata, as well as temperature, relative humidity, cloud cover,

and moisture status may influence the actual dose the plant experiences (Runeckles, 1987). However, assuming that the sensitivity of the biological target is nearly identical at both low and high elevations, some adjustment should be necessary when attempting to link experimental data obtained at low-elevation sites with air quality data monitored at high-elevation stations. Lefohn *et al.* (1990c), using Boyle's Law as a first approximation, found that the value of cumulative exposure indices that use a threshold concentration can vary substantially as a function of pressure. The authors pointed out that although the magnitude of adjustment to each hourly average concentration was less than 20%, the cumulative effect of applying a number of small corrections, that were biased in the same direction, to an exposure index with a threshold, resulted in potentially large adjustments.

Responding to Lefohn *et al.* (1990c), Larson and Vong (1990) discussed the limitations of the use of O_3 concentration as mass per unit volume and derived a correction for temperature and pressure changes. The authors' approach was based upon a theoretical evaluation of the O_3 flux relative to its value at standard conditions. Temperature and pressure were allowed to vary but wind speed and tree dimensions were held constant. The relative change in the flux to the intercellular air space of the foliage at the top of the forest canopy was estimated as a function of temperature and pressure. Larson and Vong (1990) subdivided the overall resistance to mass transfer into three components: the stomatal resistance, the needle/branch boundary layer resistance, and the turbulent air resistance at the top of the canopy. Based on their theoretical calculation, the authors concluded that if identical O_3 mass concentrations were measured at two sites separated by 2000 m elevation, the O_3 flux at the lower site would exceed the flux at the higher site by 4-

8%, due to temperature and pressure effects on both air volume and O_3 deposition velocity.

Larson and Vong (1990) considered deposition instead of concentration in their analysis. Although, at this time, there is no parameter that links deposition with direct vegetation effects, if an index were developed and exhibited cumulative properties with a threshold deposition value, an adjustment, perhaps similar to the magnitude described by Lefohn *et al.* (1990c), still appears to be required. Using the range of adjustments suggested by Larson and Vong (1990), we explored the effect on cumulative indices by applying adjustment in the range of 1-10% to each of the hourly average concentrations. We compared the values of the cumulative indices, SUM06, SUM07, and W126, after the hourly concentrations were adjusted, with the values of the cumulative indices that were based on unadjusted hourly average concentrations.

The results of the comparison are provided in Tables 2-9 through 2-11. The tables summarize the percentage differences between adjusted and unadjusted integrated exposures as a function of the percent changes made to each of the hourly average concentrations. *A review of the tables shows, depending upon the adjustment factor selected, that substantial differences occur between the unadjusted and adjusted cumulative index.* For example, in 1987, for the high-elevation Whiteface Mountain 1 site, if the effect of temperature and pressure resulted in an adjustment factor of 10%, a 47.9% reduction in the SUM07 cumulative value would have been experienced (Table 2-10). Therefore, assuming that the exposure-response models were developed using hourly average data reported in parts per million at a low-elevation location, the concentration information would have to be changed to absolute

concentration units (e.g., micrograms per cubic meter) and adjustments made to reflect the lower exposures experienced at the high-elevation site.

Additional experimental research is required to clarify this important issue (Lefohn *et al.*, 1990c; Lefohn and Lucier, 1991). Neither target sensitivity nor temperature considerations were integrated into the adjusted cumulative exposure values described above. As indicated above, temperature is not considered an important ameliorating factor when actual ambient temperatures are used. However, the sensitivity of the target organism may be an important consideration. Unfortunately, the relationship of target organism sensitivity to O_3 and to elevation and temperature has not been evaluated. In the absence of such information, the biological consequences of high-elevation exposures to the reduced absolute concentration of O_3 , that are disguised by the use of mole fraction units of concentration, need to be further investigated. For biological purposes, concentration should be converted to micrograms per cubic meter units, based upon ambient pressures and temperatures. Only then will it be possible to compare the results of biological investigations conducted at low- and high-elevation sites, particularly if these involve the generation of exposure-response relationships based upon cumulative threshold-sensitive indices.

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