Critical Levels as Applied to Ozone for North American Forests

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Abstract—*The United States and Canada have used concentration-based parameters for air quality standards for ozone effects on forests in North America. The European critical levels method for air quality standards uses an exposure-based parameter, a cumulative ozone concentration index with a threshold cutoff value. The critical levels method has not been used in North America, but efforts are now being initiated in the United States and Canada to use the critical loads concept for nitrogen and sulfur deposition to forest and aquatic ecosystems. It is expected that if this effort is successful, the critical levels approach might also be used for ozone effects on forests in North America. Although the European exposure-based or the newer flux-based critical levels standard may seem to be an improvement over the concentration-based United States and Canadian standards in relating plant response to ozone, the most appropriate ozone critical level index for plant response includes a flux-based parameter that incorporates plant defensive mechanisms. Forest plots and monitoring protocols are operational in Europe to obtain data used in determining critical levels for ozone. Similar plots should be established in the United States and Canada to begin collection of field data needed for determining critical levels for ozone in North America. The plots should be established in coordination with current Forest Health Monitoring (FHM) plots in the United States and long-term forest health plot networks in Canada. Current North American ozone monitoring networks should be expanded to obtain initial ozone data for these forest health monitoring plots.*

Ozone and Plant Response

Ozone is recognized as the air pollutant most harmful to vegetation (de Vries and others 2003, Krupa and others 2001, US EPA 1996). Yet it has been difficult to identify metrics that relate ambient ozone exposure to plant response. Active monitoring using EPA equivalency protocols for continuous ozone monitoring summarizes continuous data observations as hourly averages. Since almost all of the routine ozone monitoring of ozone in the US and Canada is recorded and/or summarized as hourly averages, it seems logical that these values have been used in the determination of a metric for the air quality standards. Various metrics of the one-hour average ozone concentrations have been used to summarize ozone to relate to vegetation response. Research has determined (Musselman and others 1994, Musselman and Massman 1999, US EPA 1996) that the ozone parameters most closely related to vegetation response are those that:

- Are cumulative throughout the growing season,
- Preferentially weight the higher concentrations, and
- Include time periods when stomata are open and ozone is taken up into plant tissue, and when the plant tissue is most sensitive to ozone.

The hourly ozone values can be used in calculation of cumulative ambient exposure of ozone to vegetation throughout a growing season. Also, the hourly ambient ozone concentration database can be used to quantify the high concentrations of ozone that impact vegetation. This is often accomplished by using a threshold value and considering concentrations equal to and above or only above that value in the index. Determination of stomatal response of vegetation requires stomatal conductance data not associated with ambient ozone concentrations but important for ozone flux into plant tissue. Stomatal conductance data are known only for a few plant species, and the data were mostly obtained from controlled environment experiments rather than from plant growing under natural conditions in the field. Thus, it is difficult to estimate ozone flux for a large diversity of vegetation and changing environmental and ozone conditions across the United States. An additional factor in the usefulness of ozone flux and its relationship to plant response is that defensive mechanisms within plant tissue must be considered in formulating a flux-based standard.

It is important to clarify the definitions of concentration, exposure, flux, and dose when discussing ozone and plant response, since these terms have been given different meanings over the past few decades (Fowler and Cape 1982, Grünhage and others 1999, Runeckles 1974). The definitions here have recently been adopted by the US EPA (2004).

- Concentration of ozone is the number of moles of ozone per unit volume of air.
- Exposure is the product of the concentration of ozone and the length of time that vegetation is exposed to that pollutant.
- Stomatal flux is the rate of uptake of ozone through stomata.
- Dose is the instantaneous stomatal flux of ozone integrated over time, or the total amount of pollutant taken into plant tissue through stomata over time.
- Effective flux is the balance between flux and defense, since detoxification mechanisms are present in leaf tissue resulting in not all the ozone being taken up being associated with vegetation injury or damage.
- Effective dose is the integral over time of the effective flux.

As one moves down this list of ozone parameter indices, the relationship of the parameter to plant response increases. Therefore, it is desirable to utilize ozone metrics for air quality standards to protect vegetation that use parameters nearer the bottom than nearer the top of the list.

Current U.S. Air Quality Standard

The United States has implemented the Clean Air Act (CAA) amendments of 1970 (Public Law 91-604) and the Environmental Protection Agency is required to promulgate primary and secondary National Ambient Air Quality Standards (NAAQS). The primary NAAQS is to "protect the public health, allowing an adequate margin of safety;" and the secondary NAAQS is to "protect the public welfare from any known or anticipated adverse effects associate with the presence of such air pollutant in the ambient air." According to the CAA, "public welfare" includes, but is not limited to "soils, water, crops, vegetation, manmade materials, animals, wildlife, visibility and climate."

NAAQS are required to be reviewed every five years. A summary document called the Air Quality Criteria Document (AQCD) is prepared based on the latest scientific knowledge and is extensively peer reviewed and open to public review. From the background AQCD document, the EPA Office of Air Quality Planning and Standards prepares an analysis (OAQPS Staff Paper) that is reviewed by an independent review board, the Clean Air Scientific Advisory Committee (CASAC). CASAC provides a "closure letter" to the EPA Administrator stating their recommendation for the ozone standard.

The first NAAQS for ozone was in 1971. Additional AQCDs for ozone have been prepared in 1978, 1984, 1993, and 1996 with subsequent NAAQS determined based on the scientific information in the AQCDs. Based on the latest scientific information in the 1996 AQCD for ozone "Air quality criteria for ozone and related photochemical oxidants" (http://cfpub2.epa.gov/ncea/cfm/recordisplay.cfm?deid=2831), the EPA prepared a OAQPS Staff Paper published in 1996 that recommended an exposure-based secondary standard of the SUM06, 12 hr/day, 3-month index based on vegetation response to ozone. This index incorporates a cumulative exposure based ozone parameter using a threshold ozone concentration, and it accounted for the time period during the day when stomatal flux is expected to be at its maximum. This recommended secondary standard was different from the recommended concentration-based primary standard.

The CASAC reviewed the 1996 AQCD and the 1996 OAQPS Staff Paper, but could not reach a consensus and "come to closure" on the form of a secondary standard for ozone. Therefore, the EPA Administrator's decision in 1996 was that the secondary standard for ozone should be the same as the primary standard; reasoning that the new more stringent primary standard for public health would provide sufficient protection for public welfare (vegetation). Thus, the current United States primary and secondary ozone NAAQS (promulgated July 1997) is 0.08 parts per million (ppm) for an 8-hr average. The standard is exceeded when the fourth highest daily 8-hr average for ozone over the last three years is greater than 0.08 ppm (0.084 effective when rounded). This United States NAAQS for ozone is not an exposure-based standard, since it is not cumulative. Nor does it preferentially weight the higher concentrations or consider stomatal uptake. Thus, the relationship between the concentration based NAAQS and vegetation response is weak. Nevertheless, improving air quality by reducing ozone to become compliant with the current NAAQS will likely reduce ozone impact on vegetation, since control strategies will likely reduce the higher hourly ozone concentrations.

For areas in non-compliance with NAAQS, the responsible local, tribal, or state air quality regulatory agencies develop "State Implementation Plans" (SIPS) which identify air pollution control strategies that will be implemented to reduce concentrations of the pollutant below NAAQS. The CAA also requires the "Prevention of Significant Deterioration" (PSD) to "protect the public health and welfare from any actual or potential adverse effect from… air pollution…not withstanding attainment and maintenance of NAAQS." The PSD section of the CAA provides protection of Air Quality Related Values (AQRVs) for certain national parks, wildlife refuges, and wilderness areas, through establishment of ceilings on additional amounts (increments) over baseline levels in these areas (called Class I). The New Source Review (NSR) process is used to evaluate the potential affect of pollutant sources on primary and secondary receptors.

Although the US has not adopted an exposure-based NAAQS, some federal agencies in the US are utilizing exposure-based parameters to evaluate ozone effects on Class I areas in their NSR analysis of ozone effects on forest vegetation, in response to the requirements of the National Environmental Protection Act (NEPA). A web based software tool (http://216.48.37.155/calculator/calculator.htm) has been developed to summarize ambient hourly ozone data to determine the W126 and N100 (as recommended by FLAG 2000), SUM06, maximum daily 8-hour averages values, percentile distributions, average concentration for each hour of the day, and frequency distributions for use in this evaluation. Using results from experimental studies in open top chambers, the W126 and N100 values are used as inputs to estimate biomass reductions of key forest species from ozone exposures.

A new External Review Draft AQCD for ozone will be available in October 2004 (http://cfpub2.epa.gov/ncea/cfm/recordisplay.cfm?deid=22411). The 2004 AQCD again documents that vegetation is very sensitive to ozone and that plant response shows a closer relationship to cumulative indices that preferentially weight the higher concentrations. The document also discusses ozone flux, effective flux, and effective dose in detail, and reviews a growing number of literature citations that now recognize that ozone taken up into plant tissue is more closely related to vegetation response than is ambient ozone concentration, regardless of how the ambient ozone concentrations are summarized (Karlsson and others 2003, Massman and others 2000, Musselman and Massman 1999). Nevertheless, the United Sates has always used a concentration basic metric to determine air quality standard for ozone, and the primary and secondary NAAQS for ozone have always been the same. Whether the forthcoming summary EPA Staff Paper and CASAC review recommends an exposure, dose, or effective dose-based secondary NAAQS for ozone that is different from the current concentration-based primary standard, and whether the EPA Administrator agrees, remains to be determined. However, changing from a concentration-based standard to an exposure, dose, or effective dose-based standard seems unlikely.

Current Canada Air Quality Standards

Canada-Wide Standards (CWSs) are established by the federal government and provincial and territorial Environment Ministers working together under the framework of the "Canada-wide Accord on Environmental Harmonization" (http://www.ccme.ca/ccme). CWSs are developed using sound scientific foundation and a risk-based approach. The CWSs developed are presented to the Provincial Ministers. The "Canada-wide Environmental Standards Sub-agreement" sets standards for the provincial governments to jointly agree on priorities to develop standards and to prepare complementary work plans to achieve those standards. Current ozone Canada-Wide Standards (CWSs) for human health and the environment, established in 2000, is 65 ppb, fourth highest daily maximum 8-hour average concentration, three-year average. Compliance is required by 2010, with a review scheduled for 2005. Note that the Canadian standard is similar to that for the United States, demonstrating that the two countries closely coordinate their ozone air quality standards. It is expected that Canadian regulators will also retain concentration-based CWSs based on ambient hourly ozone concentration data. As with the United States NAAQS, the CWSs for Canada are concentration based. They are not cumulative nor do they preferentially weight higher concentrations, and they do not consider stomatal uptake into plant tissue.

Current Europe Air Quality Standards

The European Union uses critical levels or critical loads as a basis for evaluating pollutant effects on vegetation and the subsequent setting of exceedance levels. The basis for the critical levels/loads concept is the United Nations Economic Commission for Europe (UNECE) Convention on Long-range Transboundary Air Pollution (CLRTAP) that provides international cooperation for air pollution abatement in the European Union. CLRTAP provides a framework for scientific cooperation and policy formulation and identifies specific obligation of the participating parties. Both the United States and Canada are parties of CLRTAP, but their participation has been inactive. CLRTAP established a Working Group on Effects (WGE). The WGE established a number of International Cooperative Programmes (ICPs), Working

Group Task Forces for providing a scientific basis for determination of critical levels/loads of pollutants for Europe. Critical loads are mapped by the ICP Modeling and Mapping Task Force based on scientific input from the various specialty ICP Work Group Task Forces, such as ICP-Forests and ICP-Vegetation. The information provided by the ICP Work Groups provides a sound scientific basis for the Co-operative Program for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe to formulate regulatory protocols for Europe.

European protocols use critical loads and/or critical levels to evaluate plant response to pollutants. The term critical level is generally used for gaseous air pollutants such as ozone which do not accumulate in plant tissue or ecosystems, while the term critical loads is used for pollutants such as nitrogen and sulfur which are deposited in ecosystems and accumulate over time to result in a cumulative loading on vegetation and ecosystems. Although the effects of ozone are cumulative, the pollutant itself is not cumulative in plant tissue or on non-plant surfaces. The critical loads and critical levels standards are exposure based, since they consider loading of a pollutant concentration over a period of time such as a growing season.

A critical load is defined as "the quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge" (UNECE 2004, chapter 5). Critical levels have been defined as "the atmospheric concentrations of pollutants in the atmosphere above which adverse effects on receptors, such as human beings, plants, ecosystems, or material may occur according to present knowledge (Karenlampi and Skarby 1996, UNECE 1996). Critical loads for pollutants have been defined similarly in the US by Federal Land Managers as "the concentration of air pollution above which a specific deleterious effect may occur" (FLAG 2000). More recently, the critical levels definition in Europe has been modified for vegetation to "concentration, cumulative exposure, or cumulative stomatal flux of atmospheric pollutants above which direct adverse effects on sensitive vegetation may occur according to present knowledge" (UNECE 2004, chapter 3). Note that "cumulative stomatal flux" could be called "dose" as defined above.

It is recognized that ozone is the most important regional air pollutant in Europe (de Vries and others 2003). Although the United States, Canadian, and European air quality standards all use hourly ambient ozone as a basis for calculation the ozone metric, the European critical loads concept is different from the ambient air quality standard used in the United States and Canada. The Europeans have adopted the AOT40 parameter as the index to determine critical levels for exposure of vegetation to ozone. The AOT40 is the sum of differences between the hourly ozone concentrations in ppb and 40 ppb in each hour when the ozone concentration exceeds 40 ppb. Since the AOT40 parameter is exposure based, utilizes a threshold, and is accumulated over the season for determination of critical levels, an AOT40 critical level should be more closely related to vegetation response than a concentration-only based standard like that used in the United States and Canada.

The European International Cooperative Programme (ICP) on Effects of Air Pollution on Natural Vegetation and Crops currently uses three different cumulative exposure methods to evaluate critical loads for ozone for crops, forests, and natural vegetation (http://icpvegetation.ceh.ac.uk). All three methods use hourly ambient ozone concentrations at the top of the canopy as the basis for quantifying the ozone critical loads for the evaluation. Plant response data for determining effects of critical levels of ozone are collected at or near ICP plots, or from controlled fumigation experiments. The three cumulative exposure based methods used for ozone are the Level I (AOT40) concentration based critical level, the VPD-modified concentration based critical level, and the Level II stomatal flux based critical level (UNECE 2004, chapter 3). The flux-based approach is used only for wheat and potato crops, since flux data are lacking for other crops and for forest trees and semi-natural vegetation. The concentration-based approach is used for all other agricultural crops, horticultural crops, semi-natural vegetation, and forest trees. For forest trees, the critical level for ozone is an AOT40 of 5 ppm-h, using the hourly concentration means at the top of the canopy (20m) from 1 April to 30 September. The VPD-modified concentration-based critical level is used only for injury for selected crops.

It is important to distinguish between injury and damage to vegetation from ozone. The definitions listed here have been adopted by the US EPA (2004).

- Injury is leaf necrosis, premature leaf senescence, reduced photosynthesis, reduced carbohydrate production and allocation, reduced growth, and/or reduced plant vigor.
- Damage is all effects that reduce the intended value or use of the plant. Included in this definition are reductions in economic, ecologic, or aesthetic value (US EPA 2004). Damage can be either yield loss or crop loss:
    - Yield loss is reduction in quality or quantity of the harvestable portion of a agricultural or forest crop from ozone.

- Crop loss is the reduction in the monetary value of the forest or agricultural crop due to ozone.

The European Level I assessment of ozone effects on vegetation is an exposure based analysis of injury. It is not appropriate to use the Level I assessment for yield loss, but it only should be used as an indication of relative risk of vegetation to ozone (Fuhrer and others 1997, Grunhage and others 1999, Karenlampi and Skarby 1996, Karlsson and others 2003). Level II assessment is designed to estimate effects of ozone on vegetation based primarily on ozone uptake and yield response models (Karlsson and others 2003). Stomatal flux data are used for this assessment. This dose-based critical level is beginning to be used in Europe to determine yield reductions for wheat and potato crops. However, the AOT40 exposure based critical level is now being used in Europe to determine yield loss (but not crop loss) for agricultural and horticultural crops, and for growth reductions in forest trees and semi-natural vegetation (UNECE 2004).

Although the dose-based approach would be expected to provide a better relationship between ozone and plant response and provide information to determine yield loss, limited data are currently available to implement this method. The Europeans are beginning to utilize stomatal flux and dose but not effective flux nor effective dose to determine critical levels for wheat and potato crops. Even where data are available for wheat and potato, the usefulness of this approach will be limited by lack of data on defensive mechanism that can greatly affect plant response once ozone uptake has occurred. Defensive mechanisms within plant tissue determine whether or not ozone taken into leaves is detoxified before it can injure plant tissue. Efforts have begun to quantify the defensive mechanisms in ozone plant flux response models (Barnes and other 2002, Massman and others 2000, Massman 2004, Plöchl and others 2000), but these defensive algorithms are not utilized in the current critical level models, and European efforts have concentrated on stomatal flux and dose when not using exposure-based ozone parameters, but not on effective flux or effective dose-based models.

Linking European ICP-Forests Plots and Forest Health Monitoring Plots in the United States and Canada to Critical Levels

Extensive networks of plots for forest health monitoring exist in the US, Canada, and in Europe. An extensive network for monitoring ambient ozone also exists in North America and Europe. Many of these monitors are in urban or suburban areas away from forested ecosystems, and may not be representative of ozone in remote forest ecosystems. Nevertheless, urban areas are continuing to expand in rural forested areas and the number of rural ozone monitoring stations has increased. Using statistical techniques, urban and rural ozone monitors can be used in combination to provide reasonable ozone exposure estimates across the landscape (Fraczek and others 2003, Lee and Hogsett 2001, Lefohn and others 1997).

It is important to obtain ozone data as near each forest health monitoring plot as possible, or to develop reasonable statistical estimates of ozone exposure at those sites. This is especially important in complex terrain where ozone can vary considerably over a short geographic area. Ozone monitors should be place in open areas exposed to prevailing winds to accurately determine ambient ozone levels. Samplers placed within a canopy will not be monitoring ozone to which the forest ecosystem is exposed, since ozone will already be absorbed by plant or non-plant surfaces before it reaches the sampler.

Protocols for air quality monitoring in North America and Europe require continuous monitoring, but only hourly averages of the continuous data are utilized for calculating ozone statistics for air quality standards. The hourly average ambient ozone concentrations are summarized as an 8-hr average for the concentration-based air quality standards in North American and as an AOT40 for the exposure-based critical levels in Europe. The hourly average concentrations are also utilized in the dose-based critical loads model used in Europe.

Canada has supplemented their urban active monitoring network with an extensive network of passive ozone samplers located in remote areas (Cox and Malcolm 1999). Similarly, extensive passive ozone networks have been established in California (Arbaugh and Bytnerowicz 2003) and in the Carpathian Mountains of Central Europe (Bytnerowicz and others 2002, 2004). The Carpathian network samplers were installed near forest health monitoring plots to assess ozone effects on forest health. The passive samplers are also being tested in Western Europe, and these have been installed at or near intensive monitoring plot locations (de Vries and others 2003). The advantages of the passive networks are low cost and no requirements for electric power allowing remote installation. The major disadvantages are 1) plants are sensitive to peak ozone concentrations (Musselman and Massman 1999) that are not captured in the cumulative passive ozone data, and 2) that since determination of exceedances of air quality standards and critical levels utilize hourly ozone data, passive data, which only calculates average concentration over a period of time, cannot be used to determine these exceedances.

Attempts have been made to calculate or model hourly average ozone data based on cumulative passive data (de Vries and others 2003, Krupa and others 2001, Tuovinen, 2002). It is generally understood that these models must be calibrated using active sampling data at each sampling site (Krupa and others 2001, Tuovinen 2002). This is because of the large amount of spatial (latitude, altitude, terrain) and temporal (seasonal, diurnal, meteorological) variability in ozone between sites. Relating active to passive data is particularly problematic in complex terrain. The models also do not account for the technical errors inherent with operation and analysis of the passive samplers (Tuovinen 2002). Nevertheless, some limited success has been made to determine ozone exposure dynamics based on passive sampling data when on-site calibration with active samplers and meteorological data has been deployed. The samplers can be placed in or near forest health plots to determine ozone loading at these sites. In areas where passive monitors suggest that ozone loading is high, active ozone monitors should be installed to relate forest condition and/or ozone symptomology to the dynamics of ozone exposure.

The European CLRTAP and ICP-Forests has been relatively successful in determining exposure-based critical levels for ozone effects on forest ecosystem in Europe. A basis for this success is the establishment of a network of field plots for monitoring ecosystem functioning and the effects of air pollution on the forest condition. The approach has been to describe spatial and temporal variation in forest condition (Level I), and to understand the relationship between forest ecosystem condition and air pollutants (Level II) (Haussmann and Lorenz 2004). The objective of these data is to provide information to ICP Modelling and Mapping for formulate policy to respond to air pollution stress.

The ICP-Forests network of monitoring plots has proven to be a valuable resource for determination of critical loads of nitrogen and sulphur in Europe. Consequently, Federal Land Managers in the United States and Canada are actively evaluating the critical loads approach for nitrogen and sulphur in North America. Researchers are beginning to develop critical loads for the northeast United States and southeast Canada (http://www.ecosystems-research.com/fmi/index.htm) based on model input data from various sources. It is expected that data available from ICP-Forest type plots would enhance this database. A series of demonstration plots is being explored to provide data for critical loads mapping in other regions of the United States. If these protocols are established for N and S in North America, protocols for determining critical levels for ozone could be implemented in North America. Although it appears that the United States and Canada currently will not be adopting a European Level II approach for determining ozone critical levels, the establishment of the ICP Forest monitoring plots would provide a valuable database to enable this possibility in the future.

Initial European Level I assessment of ozone-induced specific visible injury symptoms can be evaluated in or near forest health monitoring (FHM) plots if these plots provide open exposure to ambient ozone and if ozone sensitive plant species are present. Efforts have been made in the United States to establish specific plots near FHM plots that are more suitable for ozone biomonitoring, where visible oxidant stipple symptoms are recorded on up to three open grown ozone sensitive plant species. Data available from the nearby FHM plots, including biological and physiological information for individual sensitive species, and information on ecosystem structure and function, with accompanying ancillary ambient hourly ozone or extrapolated ozone data, will allow critical levels assessment of ozone effects on forest health in North America.

Air quality standards in the United States and Canada are based on concentration, and those in Europe are based on exposure with some new effort to base European standards on stomatal flux. Any effort in the United States and Canada to move beyond a concentration-based standard and beyond the European exposure and dose-based standard toward an effective dose-based standard will allow a more meaningful air quality standard for vegetation. Information provided from ICP plots and forest health monitoring plots, along with controlled fumigation experimental data, will facilitate this transition to an effective dose based standard.

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