Proposed Revision of National Ambient Air Quality Standards (NAAQS) for Ozone (2007)

(A white paper from the USDA Agricultural Air Quality Task Force [AAQTF])

Summary and Recommendations

- On July 12, 2007, US EPA (Environmental Protection Agency) proposed revisions of both primary (human health) and secondary (human welfare) National Ambient Air Quality Standards for ozone (O_3).

- A significant issue associated with the revision of the primary standard is the new EPA definition of policy relevant background (PRB) ozone concentration, since PRB has a major role in health risk assessment. AAQTF has serious concerns regarding the associated uncertainties and their consequent propagation in the risk assessment to human health. This is consistent with the views of EPA's Clean Air Science Advisory Committee (CASAC).

- Although some of the concerns regarding the uncertainties associated with the primary standard are discussed in the EPA staff paper and in the discussions below, AAQTF believes that it does not have sufficient technical expertise to provide additional comments on this human health standard.

- Thus the emphasis of this AAQTF assessment is on the form, function and the appropriateness of the exposure metrics under consideration in the proposed revision of the secondary NAAQS.

- A major concern in the context of any revision of the secondary NAAQS is that EPA has neither provided any significant additions nor any new data on the numerical relationships between ozone exposures and plant (crops and forests) responses at this time, compared to what was used in the revision of NAAQS in 1996.

- Equally important, the ozone exposure metrics (SUM06 and W126) are no different in their approach from those considered in 1996 and suffer from a lack of consistency in their performance and a lack of underlying mechanistic or process based explanatory biological reasoning.

- According to CASAC, the quantitative evidence linking specific ozone concentrations to specific vegetation effects, especially at the complex ecosystem level, must continue to be characterized as having high uncertainties due to the lack of data for verification of those relationships.

- Therefore, it is recommended by AAQTF that, given the inadequacies described in this document, at this time it would be inappropriate for EPA to establish a new

secondary standard that is different from the primary standard. AAQTF recommends that the primary and the secondary standards be the same.

- Further, EPA and USDA should initiate and provide support for scientists in academic institutions to conduct chamber-less, open field studies under real world conditions to determine the spatial and temporal trends in the effects of ambient ozone on crop productivity. Data from such studies can be used effectively in setting scientifically defensible regulatory policies. Additionally, EPA should review the research data collected to determine if there is a more appropriate exposure metric for welfare effects and consider monitoring in rural areas using methods such as passive sampling.

1. Introduction

Sections 108 and 109 of the Clean Air Act require the US EPA to establish National Ambient Air Quality Standards (NAAQS), and to review and revise them every five years if necessary. Primary NAAQS must be set at the level that is "requisite to protect the public health," allowing an adequate margin of safety; secondary NAAQS must be set at the level "requisite to protect public welfare" from "known or anticipated adverse effects." According to the U.S. Supreme Court, in setting standards that are "requisite" to protect public health and welfare, EPA's task is to establish standards that are neither more nor less stringent than necessary for those purposes.

The U.S. Environmental Protection Agency (EPA), in a Federal Register notice dated July 11, 2007 (72 FR: 37818), proposed revisions to the existing National Ambient Air Quality Standard (NAAQS) for ozone (62 FR: 38896). Currently the 8-hour primary and secondary NAAQS for ozone are met at an ambient air quality monitoring site when the average of the annual fourth-highest daily maximum 8-hour average ozone concentration is less than or equal to 0.08 parts per million (ppm), as determined in accordance with appendix I of 40 CFR part 50.

As opposed to the naturally occurring beneficial ozone layer in the upper atmosphere (stratosphere) that is formed from oxygen molecules, ozone in the lower atmosphere (troposphere) is formed in the presence of sunlight by the reactions between the oxides of nitrogen (NO_x) and hydrocarbons or reactive volatile organic compounds (VOCs), largely generated by human activity (Finlayson-Pitts and Pitts, 2000). VOCs are regulated as the precursors of ozone under NAAQS. In that context, a VOC, is defined by US EPA (40 CFR 51.100[s]), as any compound of carbon that participates in atmospheric photochemical (sunlight-driven) reactions, excluding carbon monoxide, carbon dioxide, carbonic acid, metallic carbides or carbonates, and ammonium carbonate. EPA specifically lists exempt compounds with low photochemical reactivity (lower than propane). Reactivity is also a function of the ratio of the VOCs to NO_x. Generally, higher ratios of VOCs to NO_x yield lower reactivity, but this is not true for all compounds. Nevertheless, it should be kept in mind that VOC concentrations will be a limiting factor in urban centers, while NO_x concentrations will be limiting in rural areas (Sillman, 1999). Even though some VOCs have a low photochemical reactivity (but greater than propane), and thus contribute less to ground level ozone formation compared to other more highly reactive

VOCs, the degree of photochemical reactivity is not part of the current EPA definition, as long as a compound meets its criteria for "VOC". Thus, VOCs include all organic compounds with appreciable or high vapor pressures (volatility), although vapor pressure itself is not part of the EPA's current definition. The overall characteristics of ozone production and atmospheric transport, (including its precursors), make it a regional and continental scale environmental issue. For a comprehensive discussion of the nature, production and characteristics of surface level ozone, see the North American Research Strategy for Tropospheric Ozone (NARSTO) ozone assessment documents (NARSTO, 2000).

At the global scale, agriculture is considered to contribute to approximately 50% of the total atmospheric nitrogen burden (Olivier, 1998). In the U.S. such estimates have considerable uncertainties (including those associated with emission factors for NO_x). Approximately 80 different crop species are grown throughout the US under widely differing geographies, climates, edaphic conditions and management practices. Although the majority of cultivated acreage in the US is planted with only about ten primary crops, additional uncertainties associated with NO_x emissions arise from: (1) limited data availability in rural areas as the predominant continuous measurements of air concentrations of NO_x are from urban areas (2) inaccurate estimates due to large temporal and spatial variability in trace gas composition and magnitude of trace gas emissions from agricultural activities, and (3) differing characteristics of pollutant emissions from highly dispersed animal operations.

Many crop species such as alfalfa, cilantro (coriander), cotton and tomato produce biogenic hydrocarbons (Seco, 2007; NARSTO, 2000). In addition, agricultural practices also influence the production of VOCs (including methane). Here too, there are many uncertainties due to: (1) inadequacies in measurement methods for atmospheric trace gas concentrations and their fluxes; (2) spatial and temporal sparsity of existing concentration data; (3) lack of sufficient information on sources, distribution, emissions, transport and fate of VOCs; and (4) validation of regional scale assessments.

Nevertheless, without fully knowing the extent of uncertainties associated with the current database, control measures for agricultural NO_x and VOCs could be implemented that could curtail production activities, restrict pesticide applications, designate/limit their application times and eliminate their availability, restrict animal agricultural feeding operations (emissions from ruminants and fresh waste), prescribe costly control measures for animal agriculture, and prescribe costly control measures for certain food and agricultural processing industries (including multiplier/scaling factors for VOC measurements). Also, the renewable fuels program (ethanol, biodiesel) could be greatly affected by control measures required for a more stringent standard since they too can contribute to VOCs and NO_x during fuel manufacture and subsequent use, although there is an ongoing debate regarding that question (Jacobson, 2007). Furthermore, agriculture will be indirectly impacted by stricter controls on industry. These controls result in increased costs passed on to the consumer due to special requirements for vehicles and fuels (diesel trucks and farm equipment), restrictive permitting requirements that affect industrial plant expansions, and the loss of federal highway and transit funding.

2. Primary Ozone Standard

Background:

In its staff paper (SP), EPA recommended that the standard be lowered to between 0.060 ppm and "somewhat below" the current 0.08 ppm standard to provide an appropriate degree of public health protection (U.S. EPA, 2007b). In a March 26, 2007 letter to EPA Administrator, Stephen L. Johnson, the Clean Air Science Advisory Committee (CASAC) Ozone Review Panel recommended that the primary ozone standard (to protect public health) be an 8 hour average between 0.060 and 0.070 ppm. The CASAC letter also recommended that the standard be expressed as 0.060 or 0.070 ppm (three digits after the decimal point), instead of the 0.08 ppm (two digits after the decimal point), as the current standard (Henderson, 2007). The current standard of 0.08 ppm is not exceeded with average concentrations of 0.084 ppm and below. Expressing the standard with a third digit would prevent such numerical rounding.

A significant issue associated with the revision of the primary standard is the EPA definition of policy relevant background (PRB) ozone concentration. In the final Staff Paper, policy relevant background (PRB) ozone is defined as:

> "…the distribution of O_3 concentrations that would be observed in the U.S. in the absence of anthropogenic (man-made) emissions of precursor emissions (e.g., VOC, NO_x, and CO) in the U.S., Canada, and Mexico".

This is referred to as policy relevant background since, according to the staff paper, this definition of the background facilitates separating pollution levels that can be controlled by U.S. regulations (or through international agreements with neighboring countries) from levels that are not generally controllable in this manner. Further, according to the Staff Paper "estimates of PRB O_3 concentrations cannot be derived solely from measurements of O_3, but also must be based on modeling. The global photochemical transport model GEOS-CHEM (Fiore, 2003) has been applied to estimate PRB O_3 concentrations across the U.S." The work reported by Fiore which involved modeling background ozone concentrations with all North American anthropogenic emissions of NO_x, VOCs and CO set as zero, was supported by EPA's Office of Air Quality Planning & Standards.

This is a new approach by EPA to PRB. The SP states "In the previous review of the O_3 NAAQS, the criteria document and staff paper adopted a value of 40 ppb for PRB O_3." (U.S. EPA 2007b). That value was based on monitoring data. Now, based on the modeling with GEOS-CHEM the Staff Paper states "PRB O_3 concentrations at the surface are generally predicted to be in the range of 0.015 to 0.035 ppm in the afternoon, and they tend to decline under conditions conducive to high O_3 episodes." (U.S. EPA, 2007b).

PRB plays a significant role in health risk calculations**.** Exposure and health risk analyses use the estimates of PRB. Using alternate assumptions about the background O_3

concentration greatly affects the risk estimates. For example, the SP explains, "The main reason for higher estimates in the current assessment [of O_3-related hospital admissions for respiratory illness and asthma for New York City compared to the prior review] is the use of a single value of 0.04 ppm for background in the prior review which is higher than the current modeled values . . . ." (U.S. EPA, 2007b).

The SP includes a sensitivity analysis with regard to background ozone levels. It finds, "Assuming lower PRB levels increased the number of children with a [lung function] response, while assuming higher PRB levels decreased the estimated number of children with a response." Furthermore, "estimates assuming higher PRB levels resulted in decreased estimates of non-accidental mortality incidence per 100,000 people, lower than the base case estimates by 50% or more" and "changing the estimates of PRB tended to have progressively greater impact on the estimates of mortality risk as progressively more stringent standards were examined." (U.S. EPA, 2007b).

EPA's current approach to PRB assumes that we can eliminate emissions in Canada and Mexico through negotiations. However, it is probably unrealistic to assume that the U.S., even with necessary leadership by the State Department and the involvement of other agencies could negotiate zero anthropogenic emissions from Canada and Mexico. The result is that U.S. sources will be required to reduce emissions even further to compensate for emissions originating elsewhere in North America.

CASAC addressed PRB at its meeting on August 24-25, 2006 when it reviewed the Second Draft of the Staff Paper. In its October 24, 2006 letter (Henderson, 2006), CASAC wrote:

> "The section on policy-relevant background (2.7) continues to have problems. Although the section briefly cites the results of comparison of different models and measurements, it does not adequately address the uncertainties of the global GEOS-CHEM model, and how these uncertainties are reflected in the health risk analysis. It is important to know how the PRB is related to the considered primary ozone standard and what uncertainties there are in the risk attributed to controllable sources."

CASAC again addressed the PRB issue in its letter of March 26, 2007. In that letter CASAC pointed out:

> "The Final Ozone Staff Paper does not provide a sufficient base of evidence from the peer-reviewed literature to suggest that the current approach to determining a PRB is the best method to make this estimation. One reason is that part of the PRB is not controllable by EPA."

Additional uncertainties regarding the epidemiological studies and the human health risk data used for deriving the primary standard are expressed in the Staff Paper:

- "[We] note that inherent limitations in time-series epidemiological studies raise questions about the utility of such evidence to inform judgments about a NAAQS

for an individual pollutant such as O_3 within a mix of highly correlated pollutants, such as the mix of photochemical oxidants, especially at ambient O_3 concentrations below levels at which O_3-related effects have been observed in controlled human exposure studies. We also recognize that the available epidemiological evidence neither supports nor refutes the existence of thresholds at the population level for effects such as increase hospital admissions and premature mortality." (U.S. EPA, 2007b).

- "[Additional] work is needed to better understand the influence of model specifications on the [short-term mortality] risk coefficient." (U.S. EPA, 2007b).

- "[No] clear conclusion can now be reached with regard to possible threshold levels of O_3-related effects. . . . [The] available epidemiological evidence neither supports nor refutes the existence of a threshold at the population level for effects such as increased hospital admissions and premature mortality." (U.S. EPA, 2007b).

- "Considering the importance of estimating health risks in the range of 0.04 to 0.08 ppm O_3, additional research is needed to evaluate responses in healthy and asthmatic individuals in the range of 0.04 to 0.08 ppm for 6-8 hr exposures while engaged in moderate exertion." (U.S. EPA, 2007b).

- "Most epidemiological studies of short-term exposure effects have been time-series studies in large populations. Time series studies remain subject to uncertainty due to use of ambient fixed-site data serving as a surrogate for ambient exposures, to the difficulty of determining the impact of any single pollutant among the mix of pollutants in the ambient air, to limitations in existing statistical models, or to a combination of all of these factors." (U.S. EPA, 2007b).

A similar uncertainty is expressed in the Criteria Document:

- "There exist a number of issues and uncertainties associated with the interpretation of O_3 health effects evidence in epidemiologic studies . . . . These include the use of various indices to represent O_3 exposure, exposure measurement errors (i.e., differences between ambient concentrations and personal exposure), lag period between exposure and effect, potential confounding by temporal and meteorological factors, potential confounding by co-occurrence of other pollutants, the concentration-response function, and heterogeneity of O_3 health effects." (U.S. EPA, 2006)

The USDA - AAQTF has serious concerns regarding the associated uncertainties and their consequent propagation in the risk assessment of human health. However, AAQTF believes that it does not have sufficient technical expertise to provide additional comments on this human health standard

3. Secondary ozone standard

Background:

EPA's proposed revision of the secondary NAAQS included the consideration of two different O_3 exposure metrics: SUM06 and W126. W126 was chosen at the end as one of the two alternatives, the other being a secondary NAAQS equal to the primary and any consideration of SUM06 was deleted (FR 72: 37818). These preliminary decisions of EPA require scrutiny.

First, SUM06 is defined as the sum of all hourly concentrations equal to or above 60 ppb (0.060 ppm) during a 3-month crop (plant) growth season. W126 is a sigmoidal weighting function of hourly ozone concentrations with an inflection point at 67 ppb (0.067 ppm) and giving equal weight to values above 100 ppb (0.10 ppm). This is a complicated definition of exposure metrics and response and the SUM06 and the W126 numerical terms are discussed further in the following sections.

Exposures to relatively high ambient O_3 concentrations (e.g., $\geq$ 80 ppb) for a few hours can result in symptoms of injury to leaves of sensitive plant species (Krupa, 2001). This can be represented by a short-term NAAQS. Of greater concern, however, is the chronic exposures over the whole growth season (exposures to relatively low hourly concentrations, e.g., ~40 ppb, with random, intermittent episodes or high concentrations on one or more days) that can result in adverse effects on growth, productivity or yield and nutritive quality of the plants.

The relationships between O_3 stress and plant responses are fundamentally random by their nature. The present and proposed secondary NAAQS is based on the air concentrations of O_3. Here, it is important to note that a predominant number of EPA's O_3 measurement sites are in urban; not rural agricultural or forested areas. EPA recognized this and thus, there has been a heavy reliance on modeling, without fully defining the level of uncertainty associated with the results (U.S. EPA, 2007a). For example, because there is a greater density of O_3 measurement sites (keeping in mind that a predominant number of them are urban) in eastern US compared to the west, the size of the spatial grids (km^2) used for modeling the ambient O_3 exposures is smaller (12km X 12 km) in the east compared to the west (36km X 36 km). This, consequently, results in differing spatial uncertainties. Clearly that is not a good practice, since ozone is a secondary pollutant that has large geographic scale impacts. Ambient O_3 concentrations exhibit significant temporal and spatial variability. While at sites within the so-called "boundary layer" (generally below ~1,500 m above the sea level, where the surface influences the properties of the air above, such as its temperature, wind flow etc.), hourly O_3 concentrations exhibit a sigmoidal ("S" shaped pattern, a basis for W126). Above the boundary layer the hourly O_3 concentrations exhibit essentially a flat profile (Krupa, 2001). Equally important, O_3 concentrations measured at a particular height, for example, 10 m, above the ground level must be transferred by atmospheric diffusion to the leaf surface for uptake. It is this "absorbed dose" that results in an effect (Grünhage, 1994).

In deriving the proposed secondary NAAQS, EPA has relied heavily on plant growth and yield response results obtained through experiments using above ambient, artificial O_3 exposures in open-top (OTC) chambers. In a very thorough study for his doctoral dissertation, Jetten (1992) concluded that the exposure-response curves derived under OTC conditions cannot be used directly to estimate a quantitative effect under field conditions. In the OTC studies, O_3 was the only parameter that was varied (univariate), while the influence of other air pollutants, air temperature, soil moisture, etc were kept relatively constant or largely minimized. Overall those conditions have no similarity to the real world.

Furthermore, in the preoccupation to demonstrate the deleterious effects of O_3 on plants, experimental designs were usually constructed to optimize exposure doses above an accepted or perceived level, with a description of only a part of the total response surface or ranges of response (Calabrese, 1997). Rawlings (1988) argued:

> "There are good statistical reasons for using a dose-response approach with dose (exposure) levels well above the region of immediate interest. With respect to using higher than ambient dose levels, a concern has been raised that high levels may trigger a biological process different from that causing the yield reductions at ambient levels, and as a consequence the response equations using higher dose levels would not be reliable for prediction at the low dose levels. It is certainly reasonable to expect more than one biological process to be involved in the plant response to O_3."

However, according to Rawlings (1988), with respect to EPA's National Crop Loss Assessment Network chamber data from the 1970s-1980s, none of the response equations or model outputs showed discontinuities that suggest any abrupt change in the processes. While this is true in a statistical sense, mechanistic changes in biological systems will be gradual and are dependent on the progression of the short-interval dose levels. Equally important, under ambient conditions, any observed plant response is a product of the interactive effects of many growth-regulating variables and not O_3 alone.

According to Jordan (1988), the lack of correlation between vegetation exposure statistics has posed a major problem for those trying to assess the effects of ambient O_3 exposures. The exposure indices of SUM06 and W126 proposed by EPA are not new and were considered during the previous review of NAAQS in 1996. Böhm (1991, 1995) concluded that univariate statistic such as SUM06, was unable to identify up to 39% variance in the ambient daily O_3 data matrices from 17 sites in the western United States. Neither SUM06 nor W126 has a true biological meaning. Here, it is assumed that a composite, statistically computed, seasonal, sigmoidally weighted daily, hourly ozone concentrations is similar to a single, seasonal, non-statistical, sigmoidally shaped plant growth curve that is of a different time scale and property. In that context, it is important to note that different growth stages of a crop will respond differently to the same or differing ozone exposures.

SUM06 involving a single threshold value is scientifically inappropriate in ecological effects research, as noted by two renowned ecologists, Woodwell (1975) and Odum (1979). Threshold values will vary with the plant species and within the same plant species under different growing conditions with interactions among physical, chemical and biological factors. It is a product of a stochastic process. In contrast, W126 does not involve a single threshold value, but is a sigmoidal weighting function that is particularly sensitive at mid-range concentrations, but assigns an equal weight to all hourly concentrations >100 ppb. Furthermore, the efficacy of W126 is dependent on the number of hourly ozone concentrations >100 ppb. In addition, dial hourly O_3 concentrations do not always follow a sigmoidal pattern (Krupa, 2001). Böhm (1991) described 17 different site-specific daily patterns. Furthermore, in applying W126, it is assumed that air concentrations of O_3 and the patterns of their flux to the crop canopy and uptake (absorbed dose by the crop that results in the observed response) are always in synch. In contrast, Grünhage (1997) observed that the diurnal occurrence of high hourly O_3 concentrations in the air do not always coincide with periods of optimal uptake by plants at several sites and consequently the sigmoidal weighting function can not adequately capture the importance of the O_3 concentrations occurring during periods important for crop or plant biology (diurnally and seasonally).

Discussion:

In summary, visible O_3-induced foliar injury has been reported for a number of cultivated and native plant species under ambient conditions, frequently following acute exposures. That provides a rationale for establishing a short-term (less than 8 hour) secondary NAAQS by EPA. However, foliar injury may or may not result in growth and yield reductions. Conversely, under chronic exposures, reductions in growth and yield can occur with or without foliar injury symptoms. Our knowledge of these chronic responses is derived largely from univariate, chamber studies (e.g. from the US National Crop Loss Assessment Network or NCLAN) conducted during the 1970s and 1980s and exposure-response curves derived thereof. EPA has relied heavily on the NCLAN results. The problems with these assumptions are: (1) Plants vary in their response to O_3 at the genus, species and cultivar or provenance levels. It is highly questionable whether dose-response curves developed with crop cultivars of 1980s can be used with the present day cultivars. (2) It is highly questionable whether results from chamber-based, univariate O_3 studies can be extrapolated to chamber-less, ambient conditions. In the real world, the characteristics of plant growth and productivity are a product of the joint effects of a number of highly variable abiotic (soil moisture, air temperature, air pollutants etc.) and biotic (cultivar, pathogens, pests, management practices etc.) variables. That forms the basis for the randomness of the cause and effect, both in geographic space and in time observed in the real world. EPA has not convincingly addressed either of these issues and the associated uncertainties. While addressing these concerns might appear to be daunting, there are ways to respond to the questions. EPA simply has not taken advantage of the progress in science since the last review of the ozone NAAQS in 1996.

With regard to the current proposal, in lieu of the very significant absence of measured O_3 air quality data at non-urban sites, an elegant and sophisticated, multi-step modeling

approach has been used by EPA in defining a national-level O_3 exposure surface and consequent plant responses. However, in that modeling, the error terms at each step have been propagated to the next level, until the end point was reached. EPA identified the sources of the error terms in the model and the associated uncertainties. However, no effort was made to quantify those uncertainties. Furthermore, at this time there is no mechanism to validate the results. Those are critical and major pitfalls in developing a chronic exposure secondary NAAQS.

In summary, it is implicit that any proposed secondary NAAQS should protect both managed (e.g., crops) and native ecosystems from the adverse effects of acute and chronic exposures to ambient O_3. Of particular concern are the chronic effects. It is understood, that at this time proposing a separate plant growth season-long exposure metric to account for chronic vegetation effects as a secondary NAAQS is both tenuous and controversial due to a lack of sufficient real world data. Proposing an 8-hour secondary NAAQS that is same as the primary 8-hour NAAQS is preferable until the needed data are obtained regarding chronic welfare effects in real world conditions.

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