Comments on the First Draft of the EPA Integrated Scientific Assessment Document
(External Review Draft, EPA/600/R-19/093, September 2019)
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Disclaimer: Dr. Lefohn's comments contained within this document are his own; he represents only himself; no person or organization has seen these comments prior to submission to the Government Docket; and he has not been reimbursed for the time necessary to produce these comments. His comments are directed at clarifying the state-of-science evidence that is used in the First Draft of the EPA Integrated Scientific Assessment Document. His comments are submitted into the Ozone Docket (No. EPA-HQ-ORD-2018-0274) for the purpose of providing scientific clarification to the draft ISA (EPA, 2019a) for the ozone (O_3) rulemaking activity.

About the Author

Dr. Allen S. Lefohn is currently President and Founder of A.S.L. & Associates, LLC (http://www.asl-associates.com/) in Helena, Montana. He received his Bachelor of Science degree from UCLA in 1966 and a Ph.D. in physical chemistry from the University of California Berkeley in 1969. His Ph.D. advisor was Professor George C. Pimentel, who served as deputy director of the National Science Foundation from 1977 to 1980. Dr. Lefohn, for the period 1989 – 1999, served as an Executive Editor of the internationally recognized journal *Atmospheric Environment* and is an Emeritus Editor of the Journal. Dr. Lefohn has published approximately 125 peer-reviewed publications, edited four books, presented numerous oral papers, and participated in panel presentations. He is the editor and author of the popular book *Surface-level Ozone Exposures and Their Effects on Vegetation* published by Lewis Publishers, Inc., Chelsea, MI. During a career spanning over 50 years, his research has focused on (1) developing exposure-response relationships and indices that describe the effects of O_3 on vegetation and human health, (2) investigating biological mechanisms that define the nonlinearity (i.e., weighting of the higher concentrations more than the mid- and low-level values) response to O_3 for both human health and vegetation, (3) understanding the importance of background O_3 in relation to ambient concentrations, and (4) integrating results from the EPA's air quality database for (a) characterizing co-occurrence patterns of criteria air pollutants under ambient conditions, (b) characterizing O_3 trend patterns, and (c) designing research experiments that utilize realistic ambient exposures for assessing human health and vegetation effects. He designed the hour-by-hour concentration regimes for the Adams (2003, 2006a) experiments used in Dr. Adams' controlled human exposure studies. Dr. Lefohn designed, with the assistance of Dr. Milan Hazucha of the University of North Carolina, Chapel Hill, the hour-by-hour exposure regimes for the controlled human exposure study performed by Schelegle et al. (2009), which the EPA

focused on for the current 70 ppb O_3 primary standard. He served as Chairman of the Science Advisory Committee of the Center for Ecological Health Research, University of California, Davis and served as a member of the Committee until January 2002. His research results associated with identifying exposure metrics for assessing effects have been used by the EPA. Between 2007 and 2015, EPA staff and CASAC considered the application of an exposure metric, the W126 exposure index, as the federal secondary standard to protect vegetation. Dr. Lefohn introduced the metric into the peer-review literature in 1987 and 1988. Currently the EPA uses the 8-h O_3 standard as a surrogate for the W126 index to protect vegetation; the EPA continues to use the W126 metric in the most current O_3 rulemaking activity as an indicator of the potential risk of ambient O_3 exposures to vegetation. He was the lead consultant scientist for the EPA in authoring the air quality characterization chapter and the vegetation exposure-response section for the Ozone Criteria Document in 1996 and contributed to the Ozone Criteria Documents in 1985 and 2006. Dr. Lefohn presented testimony in March 2015 to the House Committee on Science, Space, and Technology about background O_3. In 2015, Dr. Lefohn was a co-guest editor for the *Atmospheric Environment* special issue: Observations and source attribution of O_3 in rural regions of the Western United States. Dr. Lefohn is currently on the Steering Committee of the international research effort, Tropospheric Ozone Assessment Report (TOAR). Dr. Lefohn was the lead author (with 23 additional co-authors) of the well-cited TOAR paper, **Global Ozone Metrics for Climate Change, Human Health and Crop/Ecosystem Research**, which was published in April 2018 and is available at https://www.elementascience.org/article/10.1525/elementa.279/ . Dr. Lefohn is a member of the AAAS. For many years, he served as an Adjunct Professor of Environmental Engineering at Montana Tech in Butte, Montana. Dr. Lefohn continues to be an active contributor to his research areas of interest.

Executive Summary

1. There are two key fundamental principles that are important in the O_3 rulemaking activity. The first fundamental principle is Higher Hourly Average O_3 Concentrations Should be Weighted More than Middle and Lower Values when Assessing Human Health and Environmental Effects. This principle is based on the results from the Hazucha and Adams series of human health clinical studies. The result of this principle is that Haber's Rule ($C \times t = k$, where C is the concentration of the gas (mass per unit volume), t is the amount of time necessary in order to produce a given toxic effect, and k is a constant) is **not** applicable for O_3. (Section 2.1)

2. The second fundamental principle is that Daily Maximum Hourly Averaged O_3 Concentrations Will Remain Well above 0 Parts per Billion (ppb) Even if all Anthropogenic Emissions Were Eliminated Worldwide. In other words, as emissions are reduced, the full spectrum of hourly average O_3 concentrations do not shift downward toward 0 ppb. This principle is supported by observations using empirical data and atmospheric chemistry/meteorological modeling results. (Section 2.2)

3. In controlled human health clinical studies, greater O_3 peak responses have been observed in step-wise and triangular (smooth increases and decreases in concentration) exposures rather than in constant concentration exposure protocols. (Section 2.1)

4. EPA recognizes the importance of the higher O_3 concentrations by implementing its Air Quality Index (AQI) that is reported across the US. Ozone pollutant specific sensitive groups are separated by 8-h daily maximum O_3 concentrations as shown below in Fig. ES-1. The higher the O_3 concentration exposures, the greater the potential effect on human health. (Section 2.3)

8-hour Ozone Concentration	Air Quality Index Levels
0 – 54 ppb	Good (Green)
55 – 70 ppb	Moderate (Yellow)
71 – 85 ppb	Unhealthy for Sensitive Groups (Orange)
86 – 105 ppb	Unhealthy (Red)
106 – 200 ppb	Very Unhealthy (Purple)
>200 ppb	Hazardous (Maroon)

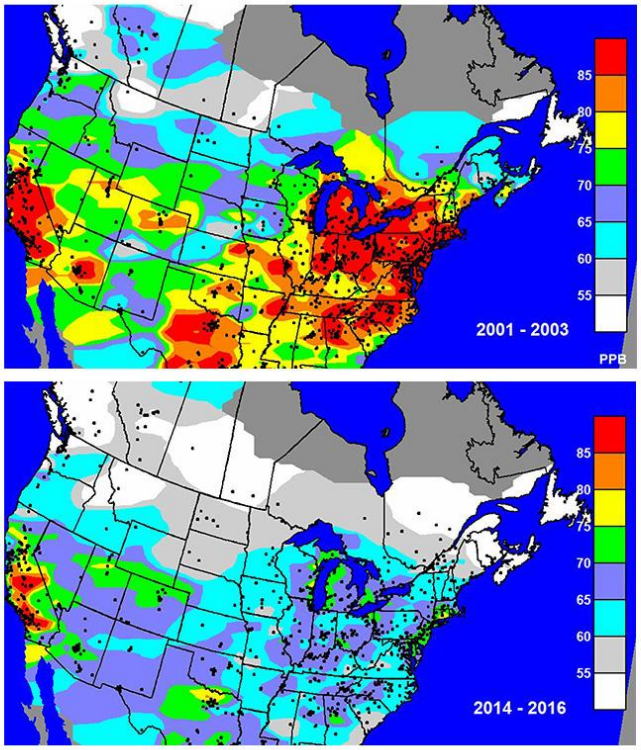
Figure ES-1. Air quality index levels (AQI) related to 8-hour concentrations.

5. During the 2015 O_3 NAAQS rulemaking (Federal Register, 2015 – page 65358), EPA noted that both acute and chronic effects could be reduced by reducing the higher hourly average concentrations. As emissions are reduced, the higher part of the distribution of hourly average concentrations moves downwards toward the middle part of the hourly average concentration distribution. (Section 2.4)

6. For vegetation, EPA (2013, 2019a) continues to conclude that (1) O_3 effects in plants are cumulative; (2) higher O_3 concentrations appear to be more important than lower concentrations in eliciting a response; (3) plant sensitivity to O_3 varies with time of day and plant development stage; and (4) quantifying exposure with indices that accumulate the O_3 hourly concentrations and preferentially weight the higher concentrations, improves the predictive power of exposure/response models for growth and yield, in comparison with using indices based on mean and other exposure indices. Results from a "natural" experiment site in the San Bernardino National Forest should be included in the draft ISA as providing independent confirmation of the experimental studies of the greater importance of the higher hourly average O_3 concentrations in influencing vegetation effects. (Section 2.2)

7. Much work has been published in recent years, particularly in Europe, using flux models for O_3 assessments at the regional, national, and European scale. There remains much unknown about O_3 stomatal uptake in vegetation at larger scales and how much uptake results in an injury or damage, which depends to some degree on the amount of internal detoxification occurring with each species. The interaction between O_3 and plant tissues is driven mainly by three distinct processes: changes in external O_3 concentration, O_3 uptake, and O_3 detoxification. The diurnal pattern of detoxification does not necessarily match the diurnal patterns of external O_3 concentration and O_3 uptake (Heath et al., 2009; Wang et al., 2015; Dai et al., 2019). I would suggest that these references be included in the draft ISA. (Section 2.2)

8. As a result of emission reductions to attain reductions in O_3 exposures, important changes occur within the distribution of the hourly average O_3 concentrations. As noted above, the highest O_3 concentrations are reduced, but other changes are also occurring, as will be discussed below. In the US, significant reductions in O_3 levels have been experienced. Figure ES-2 below compares the 3-year average of the annual 4^{th} highest 8-h value between 2001-2003 with 2014-2016. (Section 3.1)



Comparing the 3-year average of the annual 4th highest 8-h value for 2001-2003 versus 2014-2016. Analyses provided to AS Lefohn by Tom Dann, Canada. **A.S.L. & Associates, Helena, MT**

Figure ES-2. A comparison of the 3-year average of the annual 4th highest 8h-value for the period 2001-2003 with 2014-2016.

9. As emissions are reduced, models and empirical data indicate that a compression of the range of hourly average O_3 concentrations occur at many sites in the US. The higher individual 8-h daily maximum (MDA8) values are reduced toward the center of the distribution. In addition, the lowest MDA8 values ***increase***. The shifting of the lower concentrations toward the mid-level values is associated with less scavenging by NO_x of the lower hourly average concentrations as reduction in NO_x emissions occurs (Lefohn et al., 1998; U.S. EPA, 2014b; Simon, 2015; Lefohn et al., 2017, 2018). The shift of the lower hourly average O_3 concentrations toward the mid-level values indicates that the reduction of O_3 precursors results in both the high and the low concentrations shifting toward the mid-level values. ***With the shift upwards from the lower values to the mid-range values, daily maximum hourly averaged O_3 concentrations will remain well above 0 parts per billion (ppb) as emissions are reduced.*** (Section 3.1)

10. The compression of the distribution of hourly average O_3 concentrations with both the high end of the distribution and low end of the distribution shifting toward the mid concentration values results in annual average or median concentration values ***increasing*** at some sites. For example, in Fig. ES-3 below, the annual 4th highest daily maximum 8-h

concentration for 2000-2018 illustrates the trend patterns in O_3 concentrations for five sites. As expected, the southern California and the New York sites show declines over time in the 8-h metric. The three rural National Park sites at Denali National Park (AK), Voyageurs National Park (MN), and Yellowstone National Park (WY) experience O_3 exposures that are lower than the two urban sites. When the annual average is plotted (Fig. ES-4) for the same time period with the same data, the ordering of the sites from the highest to the lowest annual average concentrations appears to be counter intuitive. While the Simi Valley site in southern California experiences the highest 8-h average O_3 exposures of the 5 sites, the annual average concentration for the southern California site is comparable to values for the three rural National Park sites. The New York site experiences the lowest exposures when the annual average metric is used. Clearly, the ordering of the sites from highest to lowest exposures observed when using the 8-h metric is much different than when the annual average index is used. The increase in the annual average concentration values is associated with the low end of the distribution increasing due to less scavenging by NO_x as emissions are reduced. The three National Park sites in the figures do not experience high 8-h average concentration values comparable to many of the urban sites in the US. The high-elevation Yellowstone National Park site experiences much higher annual average values than any of the remaining 4 sites. The hourly average O_3 concentrations experienced at Yellowstone National Park (WY) are influenced by frequent occurrences of stratospheric tropospheric transport to the surface (STT-S), which is a naturally occurring process that contributes to background O_3 levels (Lefohn et al., 2001, 2011, 2012, 2014). When nonparametric statistics are applied, no trend has been observed at Yellowstone National Park using the 4^{th} highest maximum 8-h average concentration metric. (Section 3.1)

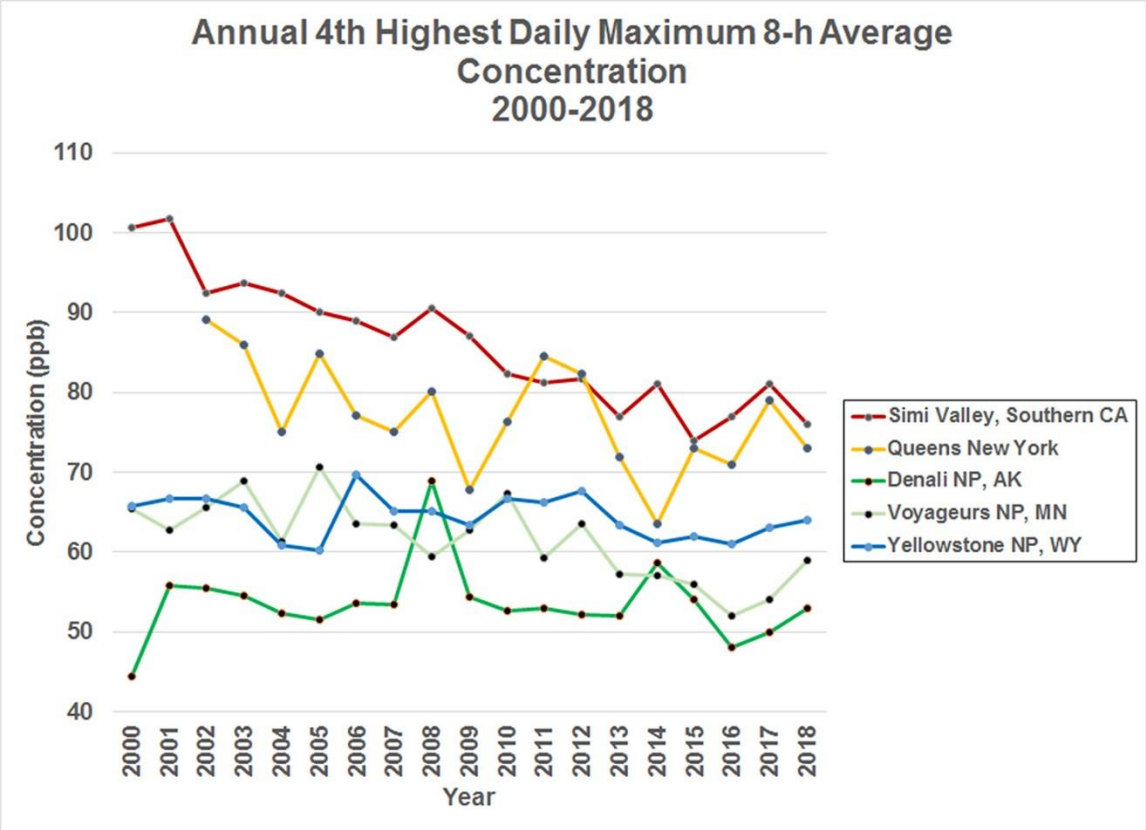


Figure ES-3. The annual 4th highest daily maximum 8-h average O_3 concentration for the period 2000-2018 for Simi Valley, CA (061112002), Queens New York, NY (360810124, Denali National Park, AK (020680003), Voyageurs National Park, MN (271370034), and Yellowstone National Park, WY (560391011).

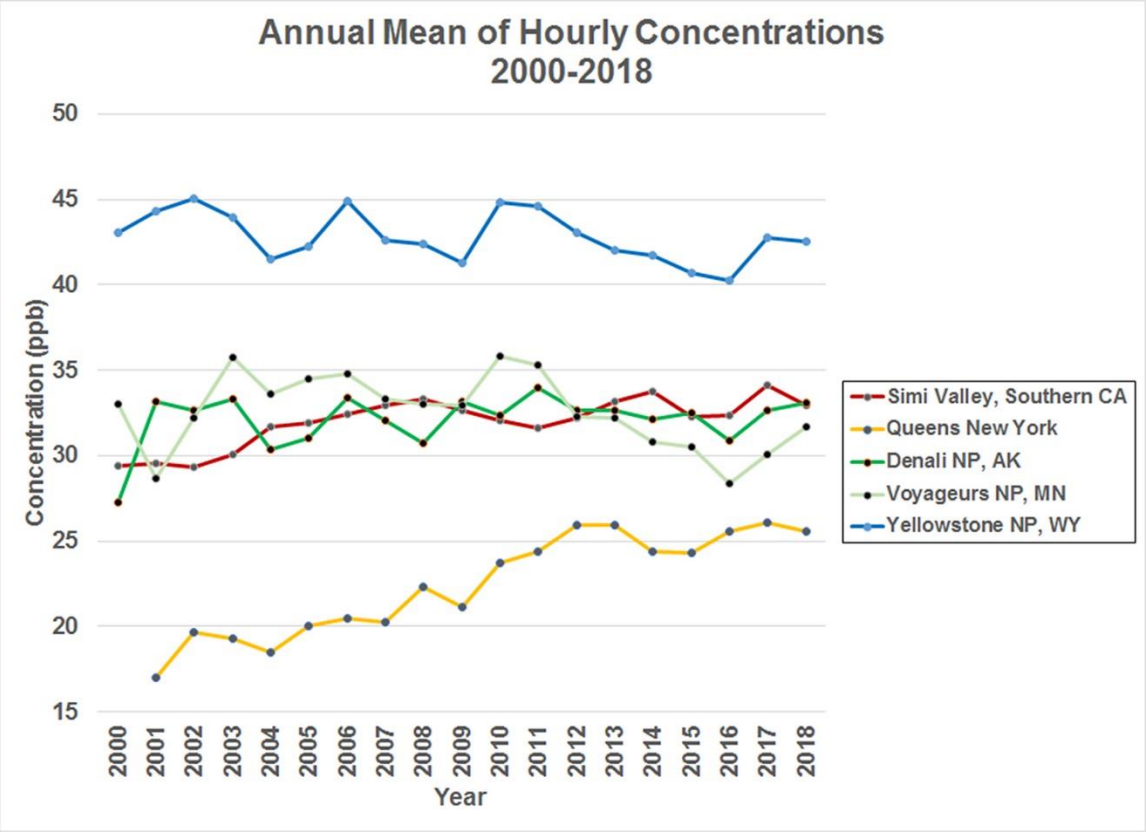


Figure ES-4. The annual mean of the hourly average O_3 concentrations for the period 2000-2018 for Simi Valley, CA (061112002), Queens New York, NY (360810124, Denali National Park, AK (020680003), Voyageurs National Park, MN (271370034), and Yellowstone National Park, WY (560391011).

11. If one believes that to eliminate "chronic" (i.e., long-term) effects from O_3 exposures one must decrease annual average concentrations, based on empirical data, annual average concentrations actually ***increase*** as emissions are reduced. The biologically important higher hourly average O_3 concentrations are reduced. By reducing NO_x to reduce the higher O_3 concentrations, there is a reduction in NO_x scavenging and a resultant upward shift of the lower concentrations toward the mid concentrations with the result that the annual average concentrations are increased (Lefohn et al., 1998; Simon et al., 2015; Lefohn et al., 2017, 2018). As indicated above, during the 2015 O_3 NAAQS rulemaking (Federal Register, 2015 – page 65358), EPA noted that both acute and chronic effects could be reduced by reducing the higher hourly average concentrations. (Section 2.4)

12. As emissions are reduced, models predict that the highest concentrations, which normally occur during the summer months, shift at some sites in the US from the summer months toward the April-June months. In addition, there are sites in the EPA's AQS database where maximum concentrations have shifted from summer to spring months, which confirms the predictions of the models. There are also sites across the US where the highest O_3 exposures occur in the spring independent of emission reductions. These

observations have important ramifications for assessing the validity of background O_3 modeling estimates. (Section 3.1.2)

13. At sites influenced by natural processes involving the stratosphere (i.e., STT-S contributions), there is a tendency for the highest O_3 exposures to occur during the spring months, but some sites (e.g., high-elevation sites) may experience STT-S contributions throughout the year. This also has important ramifications for assessing the validity of background O_3 modeling estimates. (Section 3)

14. In both the draft ISA and the draft PA, EPA appears to be mainly focused on estimating how much of current O_3 levels can be attributed to sources other than U.S. anthropogenic sources on days when ambient levels exceed the O_3 NAAQS. In other words, how much does background O_3 contribute on the days when ambient levels are highest? (Section 3)

15. There are other reasons for quantifying background O_3. In the draft ISA, EPA states that background seasonal and monthly means of hourly data are also included because longer averaging times are relevant for assessments of human health and ecological effects. This statement is not accurate. In many cases, assessment of human health and ecological effects are not based on longer averaging times. For the vegetation related W126 exposure index, which is a cumulative, rather than an average exposure metric, hourly average concentrations are weighted using a sigmoidal function and then accumulated over a specific period for assessing risk. Hourly average background O_3 concentrations contribute to the observed concentrations and therefore, contribute to the cumulative risk. For some human health risk assessments, daily 8-h average concentrations are used at times in a time series. Daily maximum 8-h average concentrations contain background O_3 concentrations, which contribute to the estimated human health risk. (Section 3)

16. The authors note that the term **U.S. background (USB)** is used to assess background O_3. No clear reason is provided in either the draft ISA or the draft PA why the authors chose to define background O_3 using the USB (i.e., zero-out) approach rather than other modeling methodologies. Simplicity of interpretation and consistency with previous analyses appear to be the reasons that USB rather than apportionment based USB_{AB} was used in the modeling described in the draft PA. The selection of USB rather than apportionment-based US background (USB_{AB}) may have important ramifications on the adequacy of background O_3 modeling results. (Section 3)

17. Given that the EPA has chosen to use USB rather than USB_{AB}, USB estimates will represent a quantity that will never occur in the real atmosphere (EPA, 2014a). As noted in the 2014 PA document (EPA, 2014a), sensitivity approaches (i.e., the methodology used for estimating USB) can be unreliable for evaluating mass contributions to O_3 production because of non-linearity in the chemistry. The US EPA (2014a) noted that the strength of the source-apportionment approach (i.e., USB_{AB}) is that it provided a direct estimate of the amount of O_3 contributed by each source category, while avoiding artifacts caused by non-linearity in the chemistry, which is a potential with the zero emissions (i.e., zero-out) modeling used to estimate background O_3 concentrations. The zero-out approach is suitable for determining what O_3 levels would have existed in recent

modeled years in the absence of all U.S. emissions, while the source apportionment approach (USB_{AB}) is suitable for determining the fraction of current O_3 originating from background sources in recent modeled years. A key point made in the draft ISA (page 1-52) is that the difference between USB and USB_{AB} is small in remote areas most strongly affected by USB sources, ***but can be substantial in urban areas strongly affected by anthropogenic sources that influence both production and destruction of ozone*** (Dolwick et al., 2015). The use of background O_3 estimates associated with USB (i.e., the zero-out modeling and its non-linearity chemistry problems) may result in more inconsistency than would have occurred if USB_{AB} had been estimated. An important question is: Even if inconsistencies exist, are these inconsistencies important? This will be addressed in future comments on the draft PA (EPA, 2019b). (Section 3)

18. On Page 1-55 in the draft ISA (EPA, 2019a), the authors note that the 2013 Ozone ISA (U.S. EPA, 2013) reported higher seasonal mean USB and NAB concentration estimates in spring than in summer for most regions of the U.S, and that these results are consistent with earlier modeling estimates. EPA notes that while some new results are consistent with this pattern, other results suggest that summer USB and baseline O_3 concentrations can be comparable to or greater than spring concentrations. The draft ISA did not resolve the conflicting conclusions about when seasonal mean background O_3 is greatest. I suggest that further discussion is warranted concerning the inconsistencies in the results. The draft ISA appears to dismiss the inconsistencies by indicating that seasonal mean USB metrics are less relevant for estimating USB concentrations when focusing on days with high MDA8 concentrations, as well as for understanding the role that hourly average background O_3 concentrations play in affecting human health and vegetation risk estimates. Actual O_3 monitoring data show that the highest O_3 exposures occur at National Park sites across the US during the springtime (March to mid-June). The EPA in its 2014 PA (EPA, 2014c) (Welfare Appendix, page 7A-12) provided the highest 3-month W126 values and the timeframe corresponding to those W126 exposures for the National Parks with O_3 monitors for the period 2006-2010. Several of the O_3 monitors in the National Parks experienced their highest W126 exposures during the spring months (defined as March, April, May or April, May, June) period. While the months of April, May, and June are not entirely a spring period (half of June is still spring), the time of year when the frequency of stratospheric intrusions (i.e., a natural process) to the surface is greatest at many sites during the March – June window. Besides the National Parks, there are other monitoring data where the highest O_3 exposures occur during the March-June period. I suggest that the authors of the draft ISA evaluate O_3 data in the EPA's AQS database and quantify when the highest O_3 exposures occur at various types of sites. This I believe would add to the discussion. There continues to be strong evidence, as supported in the literature, that background O_3 across the US is highest at many sites during the springtime (including into the month of June) and background O_3 is an important contributor at many high-elevation sites throughout the year. Section 3.2.4)

19. The draft ISA attributes increasing trend patterns observed at high-elevation western U.S. sites to long-range transport from Asia. There is inconsistent on the influence of long-range transport from Asia on western high-elevation O_3 monitoring sites. Long-range transport from Asia has not influenced trend patterns at all western US high-elevation O_3

monitoring sites. Not all high-elevation western U.S. sites have exhibited statistically significant trends during the springtime, when transport is expected to be highest from Asia. An evaluation of trend patterns of high-elevation western U.S. sites during the springtime, using the 4th highest daily maximum 8-h concentration exposure metric, shows that some sites do not experience increasing trends over the period 2000-2014. I suggest that the authors indicate caution in drawing the strong conclusions currently in the draft ISA document about the impact of long-range transport on western high-elevation O_3 monitoring sites, as well as anticipated changes as a result of emission reductions in Asia. Section 3.2.6)

20. Depending upon the specific monitoring sites, background O_3 contributes varying amounts to the higher hourly average O_3 concentrations. For example, the high-elevation Yellowstone National Park site in Wyoming is dominated by background O_3 throughout the year with minor anthropogenic contributions (Lefohn et al., 2014). In Fig. ES-5 below, the relative comparison of background O_3 levels (noted by blue) to anthropogenic (noted by red) within each concentration level shows that background contributes greater than 80%, including the mid-range concentrations (20-25 ppb). In comparison, Fig. ES-6 illustrates that for the Los Angeles area, a site heavily influenced by anthropogenic emissions, background O_3 contributes less than 40% at the higher hourly average concentrations. (Section 3.2.5)

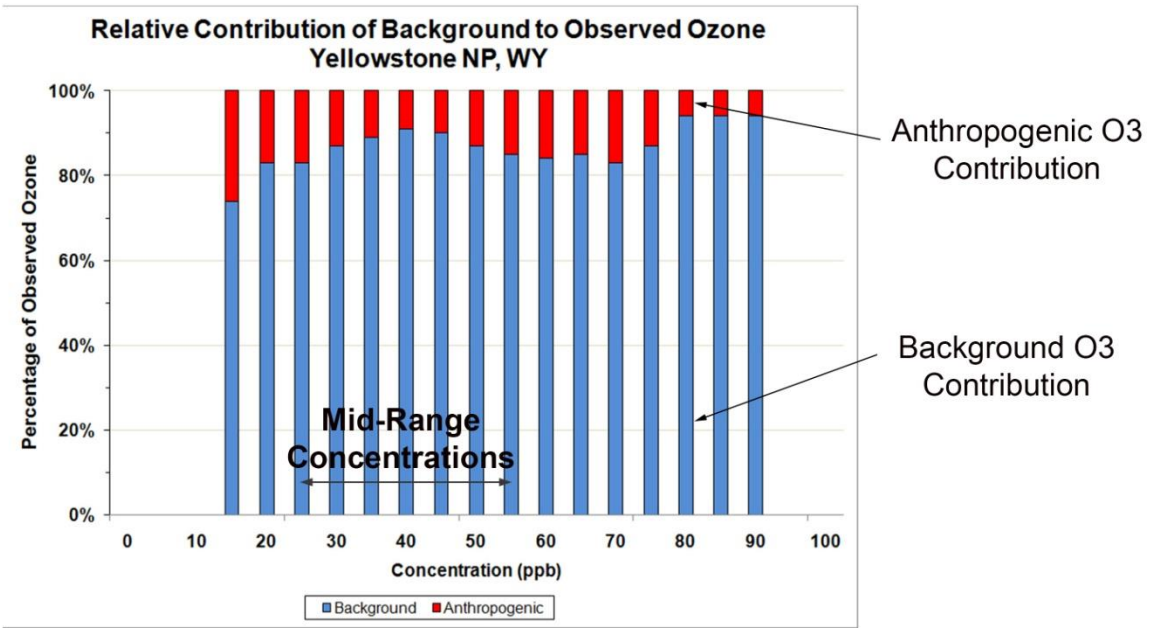


Figure ES-5. Average relative contributions of current hourly background (blue) and anthropogenic ozone (red) for Yellowstone NP (WY) (AQS ID 560391011) in 2006. (Source: Lefohn et al., 2014).

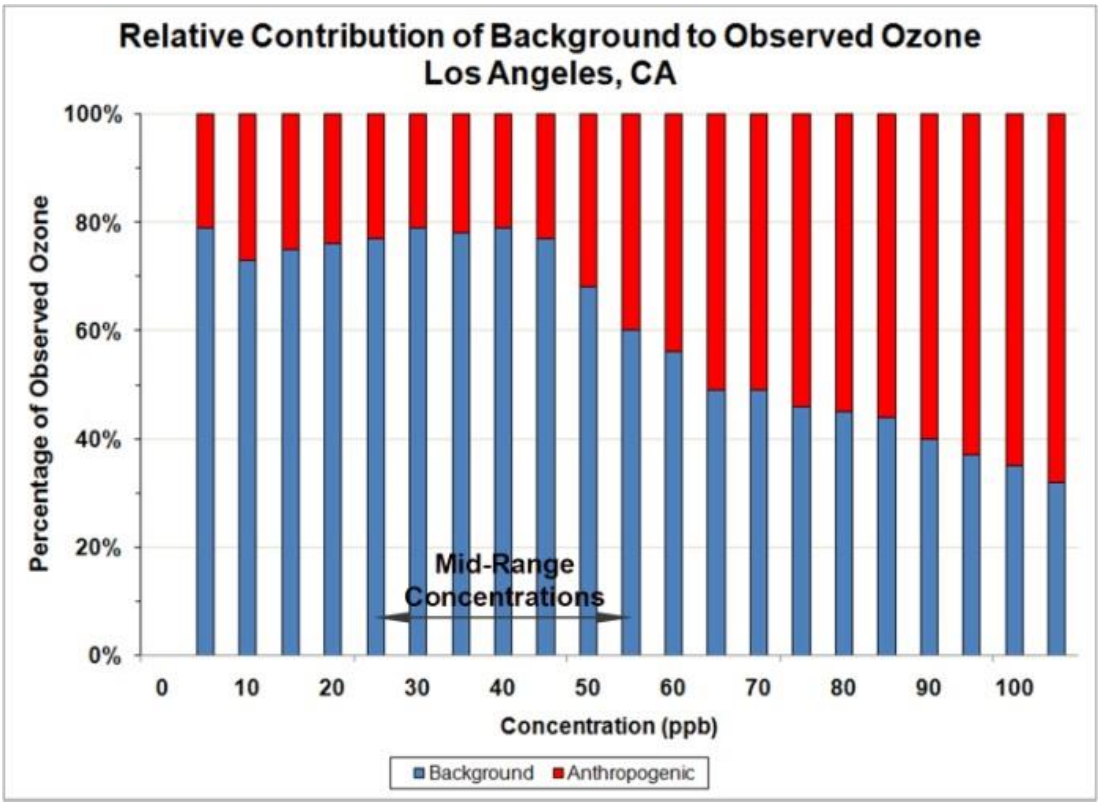


Figure ES-6. Average relative contributions of current hourly background (blue) and anthropogenic ozone (red) for Los Angeles (CA) (AQS ID 060719004) in 2006. (Source: Lefohn et al., 2014).

1. Introductory Comments

The first drafts of the EPA's Integrated Science Assessment for Ozone (ISA) (EPA, 2019a) and Policy Assessment (PA) (EPA, 2019b) documents were issued on September 25, 2019 and October 31, 2019, respectively. During the last O_3 rulemaking activities, the first drafts of the ISA, Health Risk and Exposure Assessment (HREA)/Welfare Health Risk and Exposure Assessment, and PA were issued in March 2011, July 2012, and August 2012, respectively. The final versions of the ISA, HREA/Welfare REA, and PA were published in February 2013, August 2014, and August 2014, respectively. The compressed schedule during this rulemaking cycle has resulted in the simultaneous preparation of the ISA (EPA, 2019a) and PA (EPA, 2019b). The has resulted in little time being available to integrate key concepts presented in the ISA to be properly evaluated by the general public, as well as policy makers and various stake holders (e.g., environmental groups and industry advocates). Therefore, key concepts developed in the ISA have not been exhaustively reviewed and commented upon before being integrated into the draft PA.

Accordingly, because of the compressed review schedule, my review comments, while focused on the draft ISA, also include reference to material in the draft PA. I believe it is important that the reader understand how specific issues that require further refinement in the draft ISA are currently incorporated into the draft PA. Separate to these comments, a separate set of comments will be submitted to the Docket on the PA.

1.1 Units of Concentration

In previous ISA and PA documents, O_3 metrics (e.g., annual 4th highest 8-h daily maximum average O_3 concentration and the vegetation W126 cumulative exposure index) have typically been developed using the mixing ratio unit of parts per million (ppm) or parts per billion (ppb) which, in the case of O_3, refers to the number of O_3 molecules per million or billion moist, ambient air molecules in a fixed volume. The units of the W126 metric is ppm-hrs or ppb-hrs. However, as noted in the draft ISA (EPA, 2019a), in the atmospheric science literature, ppm and ppb are not considered concentration units. Some atmospheric scientists have suggested that when referencing an O_3 observation, that the term mole fraction of O_3 in air, expressed in SI units of nmol mol^{-1} be used. To respond to this concern, the draft ISA (EPA, 2019a) states the following on page 1-3:

> Ozone concentration metrics are generally based on measurements or estimates expressed as a volume-volume *mixing ratio*, with units of parts per million (ppm) or parts per billion (ppb). Technically, ppm and ppb are not concentration units, which are defined as moles per unit volume and depend on temperature and pressure. This distinction is generally acknowledged in the atmospheric science literature. In contrast, the term *mixing ratio* is rarely used in the literature on health and vegetation effects but is instead usually substituted with the term *concentration*, understood to be more broadly interpreted as the amount of a substance in a fluid without distinguishing units. For this reason, the term *concentration* is generally used instead of *mixing ratio* in this document to

maintain consistency with its use in the health and ecological effects literature. *Mixing ratio* is still used in the more technical discussions of atmospheric sources and chemistry in Section 1.3 and Section 1.4.

I would suggest that the wording be modified. The current O_3 standard is in units of ppm. During the peer-review process of the Lefohn et al. (2018) paper, there was a suggestion by the reviewers that the use of the units ppm and ppb were not appropriate and that we should eliminate entirely the word "concentration." The reviewers suggested we use the terminology mole fraction. Some of my coauthors and I had serious concerns that the (1) use of the unit mole fraction and (2) elimination of the word "concentration" from our paper would compromise our ability to communicate with researchers in the biological area, regulators, and policymakers.

After much discussion, the authors and editor reached an agreement that the following wording, which was inserted into Section 1.2 (Ozone metrics in the context of TOAR) of Lefohn et al. (2018), was acceptable:

> In TOAR, specific units are used when describing ozone observations and levels of exposure. When referencing an ozone observation, which is measured from moist, ambient air, TOAR follows World Meteorological Organization guidelines (Galbally et al., 2013) and uses the mole fraction of ozone in air, expressed in SI units of nmol mol^{-1}. Ozone metrics (e.g., annual 4^{th} highest 8-h daily maximum average ozone value) have typically been developed using the mixing ratio unit of parts per million (ppm) or parts per billion (ppb) which, in the case of ozone, refers to the number of ozone molecules per million or billion moist, ambient air molecules in a fixed volume. In reference to units of nmol mol^{-1} and ppb, Galbally et al. (2013) states: "For all practical purposes the two quantities can be used interchangeably and without distinction". To maintain consistency with the ozone human health and vegetation research community, TOAR uses units of ppb or ppm (or ppb-hrs or ppb h for cumulative indices) when discussing ozone in terms of an exposure metric. Although the usage of the word "concentration" without specifying atmospheric conditions when referring to mole fraction (nmol mol^{-1}) and mixing ratios (ppb) is technically incorrect, the vast amount of literature on ozone health and vegetation effects uses the conventional term "concentration" when referring to an ozone level. This common usage does not distinguish between mixing ratio metrics or true concentrations metrics such as µg m^{-3}. To enhance the link to the health and vegetation effects literature and national and international policy, as well as to facilitate the understanding of this paper by health and vegetation effects scientists, the word "concentration" is used when appropriate.

I have provided the wording above as an example of the wording that might help clarify why ppb and ppm units are used in the draft ISA and draft PA.

1.2 Purpose of the Integrated Science Assessment (ISA) Document

The purpose of this Integrated Science Assessment (ISA) is to draw upon the existing body of evidence to synthesize and provide a critical evaluation of the current state of scientific knowledge on the most relevant issues pertinent

- to the review of the NAAQS for O_3 and other photochemical oxidants,
- to identify changes in the scientific evidence bases since the previous review, and
- to describe remaining or newly identified uncertainties.

The ISA's goals are to:

- Assess whether new information (since the last Ozone NAAQS review) further informs the relationship between exposure to O_3 and specific health and welfare effects.

- Provide new information as to whether the NAAQS (comprised of indicator, averaging time, form, and level) are appropriate.

An important challenge of the ISA is how to update the state of the science from that available for the 2013 Ozone ISA and place these results into perspective with key findings contained within previous O_3 rulemaking documents (i.e., 2006 AQCD for Ozone and Related Photochemical Oxidants (U.S. EPA, 2006), the 2007 Staff Paper (U.S. EPA, 2007), the 1996 AQCD and Staff Paper for Ozone and Other Photochemical Oxidants (U.S. EPA, 1996a, b), the 1986 AQCD for ozone (U.S. EPA, 1986) and its Supplement (U.S. EPA, 1986b), and the 1978 AQCD for Ozone and Other Photochemical Oxidants (NAPCA, 1978)). It is extremely important that the ISA provide its readers with a firm grasp of the foundations on which past scientific decisions were made. ***If this is not accomplished, then it is possible that the rationale for past scientific findings may be lost to history and the latest results provided greater weight than the firm scientific findings of the past.*** What is important here is that the latest results are compared to past findings and where disagreement is found, the ISA devotes space to discussing the ramifications of the disagreement. In some cases, the draft ISA has not focused on the disagreement and has not critically evaluated possible scientific reasons for the disagreement in the results. I will discuss specific examples in detail later in my comments.

I believe two key scientific fundamental principles help guide the form and level of the two Federal O_3 standards (i.e., health and welfare) in the United States. The first fundamental principle, based on (1) controlled human exposure studies and (2) laboratory and empirical vegetation experiments, states that ***higher hourly average ozone concentrations should be weighted greater than the middle and lower values when assessing human and environmental effects***. Without this key scientific fundamental principle, the selection of O_3 exposure indices for assessment purposes would be based on the personal choice of the investigator rather than based on biologically relevant metrics. Lefohn et al. (2018) discuss in detail the rationale for the selection of specific exposure metrics for assessing human health and vegetation. The second fundamental principle, linked to natural processes (e.g., stratospheric intrusions, lightning, wildfires, and soil emissions), states that ***daily maximum hourly averaged ozone concentrations will remain well above 0 parts per billion (ppb) even if all anthropogenic emissions were eliminated worldwide.*** This fundamental principle means that if global anthropogenic emissi0 parons were eliminated entirely, hourly maximum daily surface O_3 concentrations would remain

well above zero ppb and cannot attain the unreasonable goal reaching a daily maximum concentration of 0 ppb. At most locations in the world, it is not possible to attain hourly average O_3 levels on a sustained basis near 0 ppb. This second fundamental principle has profound implications for those who develop models that predict mortality and morbidity effects at O_3 levels below natural background levels. In the following comments below, I discuss the two very important fundamental principles and its relation to the O_3 rulemaking process.

2. Fundamental Principle No. 1: Higher hourly average ozone concentrations should be weighted more than middle and lower values when assessing human and environmental effects

2.1 Human Health

For the 2015 O_3 rulemaking, the US EPA noted that higher O_3 concentrations have a proportionately greater impact than lower concentrations and therefore, are an important consideration in determining ozone's health impact. Important published results by Hazucha et al. (1992) and Adams (2003; 2006a, 2006b) formed the scientific foundation for the focus on higher O_3 concentrations. The work utilized ambient-type elevated concentrations and compared them to constant concentration exposures with the same concentration × time product to assess respiratory effects. Commenting on these earlier results, Lefohn, Hazucha, Shadwick, and Adams (Lefohn et al., 2010) concluded that higher O_3 concentrations are important in FEV_1 responses and that ***the effect is cumulative*** based on exposure. EPA notes in the draft ISA (page 3-11) that greater peak responses have been observed in stepwise and triangular (smooth increases and decreases in concentration) exposures versus constant concentration exposure protocols. I suggest that the EPA include the Hazucha et al. (1992) paper in its discussion in this section because their work was one of the first to identify the importance of weighting the higher hourly average concentrations in FEV_1 response. The Hazucha et al. (1992) effort, although applying hourly values above the levels currently considered for the primary Ozone NAAQS, represents an important part of the evolution of thinking in this important research area. Conclusions of these important papers are as follows:

Hazucha et al. (1992)

> Several recent studies have suggested that in estimating exposure dose (O_3 concentration [C] × exposure time [T] × ventilation [V]), O_3 concentration needs to be weighted more heavily than either ventilation or duration of exposure in the estimates. Our observations demonstrate that the product of $\bar{C}$(mean) × T × V is not a sufficient index of exposure.

Adams (2003, 2006a)

> These results support previous evidence that O_3 concentration has a greater singular effect in the total inhaled O_3 dose than do V_E and exposure duration.

Lefohn and Hazucha (2007)

Results from controlled laboratory exposures of human volunteers indicate that higher ozone (O_3) hourly average concentrations elicit a greater effect on hour-by-hour physiologic response (i.e., forced expiratory volume in 1 s [FEV_1]) than lower hourly average values, which implies a nonlinear dose–response relationship. To date, most of the empirical models derived from laboratory human experiments of concentration dose–response have been based on a constant exposure regime. The relationship between O_3 and spirometric lung function decrements is not linear. In attempting to derive the O_3 exposure–response relationship, we urge caution in curve-fitting exercises that focus on identifying the ‘‘best-performing’’ mathematical functions. Some of the functions identified may not be physiologically relevant. We recommend that investigators focus on identifying models that have biological plausibility and apply these models with data that are derived from variable exposure regimes. The use of a logistic (i.e., sigmoid) model appears to be biologically justified. It is continuous, does not require the identification of a population threshold concentration, and deals with plateau considerations at the high end of the distribution of exposures.

Lefohn, Hazucha, Shadwick, and Adams (2010)

Controlled human laboratory studies have shown that there is a disproportionately greater pulmonary function response from higher hourly average ozone (O_3) concentrations than from lower hourly average values and thus, a nonlinear relationship exists between O_3 dose and pulmonary function (FEV_1) response. We have reanalyzed data from five controlled human response to O_3 health laboratory experiments as reported by Hazucha et al. (1992), Adams (2003, 2006a, 2006b), and Schelegle et al. (2009). Our findings indicate a common response pattern across most of the studies. Schelegle et al. (2007) introduced a concept of a phased ventilatory response associated with O_3 exposures based on the frequency of breathing (fB) endpoint. In a subsequent paper, Schelegle et al. (2009) applied this concept to include an FEV_1 endpoint. Based on VAR/STW(i.e., variable/stepwise) FEV_1 response pattern, we have used a similar approach and identified three FEV_1 phases associated with exposure to VAR/STW O_3 concentrations: (i) a 2 to 3-h initial “induction phase” in response, (ii) followed by the onset of a statistically significant FEV_1 nonlinear “response phase,” and (iii) a final “reversal phase” (i.e., change in direction of the slope of the FEV_1 decrement towards baseline as the hourly average O_3 concentration is decreased). The first phase noted by Schelegle et al. (2009) more or less coincides with our Phase 1. Their second and third phases correspond to our second phase. We have added a new third phase, the “reversal phase,” which was absent in Schelegle et al.’s (2009) study. Results from these controlled human laboratory studies applying ambient pattern exposures (Adams 2003, 2006a, 2006b; Hazucha et al., 1992) illustrate the importance of the higher hourly average O_3 exposures compared to the lower hourly average values and a nonlinear relationship between O_3 dose and FEV_1 pulmonary function (Hazucha & Lefohn 2007).

Lefohn et al. (2018)

Controlled human exposure studies that explore induced decrements in lung function indicate that the higher ozone concentrations should carry greater weight than the moderate and lower concentrations (Hazucha and Lefohn, 2007; Lefohn et al., 2010b). Such studies vary the (1) intensity, duration and frequency of exercise from light to very heavy load on a treadmill or a bicycle ergometer to increased minute ventilation, (2) duration of exposures over 6.6-h and 8-h periods, and (3) application of varying hour-by-hour concentrations versus constant concentrations. In the 1980s and early 1990s, US EPA investigators published the initial studies on the effects of 6.6-h exposures on healthy humans (Folinsbee et al., 1988; Horstman et al., 1990). In 1992, the first 8-h exposure study of ozone on lung function comparing the results using a constant concentration and variable concentration profile that mimicked typical diurnal patterns existing under ambient conditions was published (Hazucha et al., 1992). Both the constant and the variable concentration regimes used the same effective dose although the variable regime included exposure to high hourly average ozone concentrations. Compared to the square-wave exposure profile, the hourly lung function decrements in pulmonary function of subjects exposed to the variable concentration regime were substantially greater one hour after the peak exposure, with the conclusion that the higher concentrations should be weighted more than the mid- and low-level values. Several later studies (Adams 2003, 2006a, 2006b) employing either variable (continually changing) or stepwise (increasing or decreasing from one hour to the next) exposure profiles confirmed the results reported by Hazucha et al. (1992). These studies showed that equivalent doses (varying versus constant exposures) produced different responses which depended on the applied hourly ozone concentration pattern.

Page 3-11 in the draft ISA, the EPA notes:

Although greater peak responses have been observed in step-wise and triangular (smooth increases and decreases in concentration) exposures versus constant concentration exposure protocols, similar FEV_1 responses have been reported at 6.6 hours regardless of the exposure protocol (i.e., constant versus step-wise) for average ozone exposures to 60, 80, and 120 ppb (Adams, 2006, 2003a; Adams and Ollison, 1997).

Although this statement is accurate based on documenting the FEV_1 response after 6.6 hours, compared to the square-wave exposure profile, the hourly lung function decrements in pulmonary function of subjects exposed to the variable concentration regime were substantially greater one hour after the peak exposure. The fact that greater instantaneous FEV_1 decrements occurred in the variable exposure regimes means that the 8-h standard may not be as protective as intended. For the Adams (2003, 2006a) studies, I designed the hour-by-hour O_3 concentration regimes.

The hourly averaged concentrations were designed to increase in an "almost" symmetric manner. For the 80 ppb step-wise exposure, the peak hourly average concentration occurred at 4th hour and then began to decrease. The result was that the three FEV_1 phases described in Lefohn et al. (2010) (Lefohn, Hazucha, Shadwick, and Adams), associated with exposure to the variable/stepwise O_3 concentrations occurred. The first phase is the 2 to 3-h initial "induction phase" in response. The second phase is the onset of a statistically significant FEV_1 nonlinear "response phase," and the final phase is the "reversal phase" (i.e., change in direction of the slope of the FEV_1 decrement towards baseline as the hourly average O_3 concentration is decreased). Note that although the "reversal phase" occurred, the FEV_1 response remained statistically significantly different than the control at the end of the 6.6-hour experiment for several of the exposure regimes applied. The reversal phase was noted in the draft ISA and was presented as evidence supporting the statement on page 3-11 of the ISA that "…similar FEV_1 responses have been reported at 6.6 hours regardless of the exposure protocol (i.e., constant versus step-wise)." However, as noted above, compared to the square-wave exposure profile, the hourly lung function decrements in pulmonary function of subjects exposed to the variable concentration regime were substantially greater one hour after the peak exposure and may indicate that the 8-h average form of the O_3 standard may need to be revisited in the future.

What we do know from the Hazucha et al. (1992), Adams (2003, 2006a) results is that ***"controlled human laboratory studies have shown that there is a disproportionately greater pulmonary function response from higher hourly average ozone (O_3) concentrations than from lower hourly average values and thus, a nonlinear relationship exists between O_3 dose and pulmonary function (FEV_1) response"*** (Lefohn, Hazucha, Shadwick, and Adams, 2010). The implication of this is that a simple cumulative calculation of C x T (concentration multiplied by time) is not a valid determination of cumulative O_3 exposure. For understanding cumulative O_3 effects on FEV_1, in future controlled human health laboratory experiments, I believe there should be a differential weighting of the hourly average concentrations over the time of exposure, as well as a better understanding of the "induction" and "reversal" phases.

During the 2015 rulemaking O_3 activity, one controlled human exposure experiment published was the key study that the EPA focused on for the selection of the primary O_3 standard. While other controlled human exposure and epidemiological studies were also available, the EPA decided to focus on the Schelegle et al. (2009) study for setting the primary O_3 standard. Professor Milan Hazucha and I designed the hour-by-hour exposure regimes for the Schelegle et al. (2009). However, before discussing why this single controlled human exposure experiment outweighed the other studies, I would like to describe the history on the science that provides the context for why, among all the studies in the published literature, this single experiment formed the basis for the current level of the O_3 standard to protect human health.

When evaluating the epidemiological and controlled human exposure study results, that the higher O_3 concentrations were an important consideration (Federal Register (2015 – page 65354). The EPA (Federal Register, 2015 – page 65343) agreed with the conclusions of the Agency's Policy Assessment Report (EPA, 2014a) that controlled human exposure studies provided the most certain evidence indicating the occurrence of health effects in humans following exposures to specific O_3 concentrations. Specifically, the Agency recognized that the effects reported in controlled human exposure studies were due solely to O_3 exposures. In

contrast, epidemiological studies incorporate confounding factors that may have obscured the cause-effect relationship.

The EPA noted that controlled human exposure studies report the combination of lung function decrements and respiratory symptoms in healthy adults engaged in intermittent, moderate exertion following 6.6 hour exposures to concentrations as low as 72 ppb (based on Schelegle et al., 2009), and lung function decrements and pulmonary inflammation following exposures to O_3 concentrations as low as 60 ppb (based on Kim et al., 2011) (Federal Register, 2015 – page 65343). However, the EPA also noted that a level of 60 ppb is below the lowest concentration where the *combined* occurrence of respiratory symptoms and lung function decrements were observed (i.e., 72 ppb), a combination judged adverse by the American Thoracic Society according to the EPA (Federal Register, 2015 – page 65357). The EPA, noting the Kim et al. (2011) findings, has less confidence that health effects would occur below an O_3 concentration of 72 ppb. Thus, only one controlled human exposure study (i.e., Schelegle et al., 2009) informed the EPA's decision as to the level of the standard.

For the epidemiological studies, the EPA noted that the interpretation of studies' results was complicated by the presence of co-occurring pollutants or pollutant mixtures. In addition, the EPA placed less weight on epidemiologic-based risk estimates because of key uncertainties about (1) which co-pollutants was responsible for any health effect observed, (2) the heterogeneity in effect estimates between locations, (3) the potential for exposure measurement errors, and (4) uncertainty in the interpretation of the shape of concentration-response functions for O_3 concentrations in the lower portions of ambient distributions. As noted in the Health Risk and Exposure Assessment (HREA) (EPA, 2014b) with respect to the epidemiological studies, not differentially weighting the concentration-response functions at higher O_3 concentrations greater than the mid- and low-level values resulted in small differences in the estimates of mortality and morbidity risks as a theoretical effort was made to impose more stringent standards. This occurred because as shown by EPA's modeling, as well as our trends results, as emissions are reduced to meet lower standards, the high end of the concentrations shifts downward (i.e., reducing mortality) but the low end of the distribution of concentrations shifts upward (i.e., increasing mortality), ***resulting in a small net benefit***. Because of the limitations in the epidemiology studies, the EPA did not use these studies to set the proposed range for the level of the standard except for the margin of safety consideration.

As described above, the EPA (Federal Register, 2015 – page 65357) had less confidence that adverse effects would occur following exposures to O_3 concentrations below 72 ppb. With the findings of the epidemiological studies being discounted because of numerous uncertainties affecting the interpretation of the results, the EPA relied on a single study to focus on the setting of the 2015 O_3 standard. The EPA went on to conclude that a standard level as high as 70 ppb, which CASAC concluded could be supported by the scientific evidence, could reasonably be judged to be requisite to protect public health with an adequate margin of safety (Federal Register, 2015 – page 65363).

2.2 Vegetation

For vegetation, EPA reached the conclusion in 2015 that the higher concentrations should be weighted greater than mid and lower values (Federal Register, 2015 – page 65373). This conclusion coupled with the cumulative nature of the effects of O_3 on vegetation is the basis for the EPA recommending the W126 exposure index (Lefohn and Runeckles, 1987; Lefohn et al., 1988) for assessing vegetation risk.

The interest in identifying O_3 exposure regimes for eliciting adverse effects began earlier with the vegetation than the human health researchers. As indicated above, while controlled human health clinical study results regarding the importance of the higher hourly average O_3 concentrations were reported in the early 1990s (Hazucha et al., 1992), vegetation researchers reported in the 1960s that higher O_3 concentrations were an important factor for assessing vegetation O_3 effects. High O_3 concentrations were found in the 1960s to affect plant injury (e.g., spots on plants) (Heck et al., 1966). Little research on the importance of higher O_3 concentrations in relation to the mid and lower levels affecting plant damage (e.g., growth) had been performed prior to the 1980s. In December 1981, I held an informal discussion with the EPA at its research laboratory in Corvallis, Oregon. I discussed a possible hypothesis relating to the relative importance of the higher O_3 concentrations versus mid- and lower-level hourly average values for assessing plant damage resulting in economic impact. During the discussion, EPA inquired if I could design exposure regimes for the EPA that would test the hypothesis that the higher hourly average O_3 concentrations should be weighted more than the mid- and low-level values.

Soon after the December 1981 Corvallis discussion, Lefohn and Benedict (1982) published their paper that hypothesized that the higher hourly average concentrations should be provided greater weight than the mid- and low-level values when assessing crop growth reduction. In 1983, Musselman et al. (1983) provided experimental evidence supporting the hypothesis. Hogsett et al. (1985), applying exposure regimes designed by me, provided additional support to the Musselman et al. (1983) findings about the importance of the higher hourly average O_3 concentrations receiving greater weight than the mid and lower values in affecting vegetation.

Following the initial vegetation experiments by Musselman et al. (1983) and Hogsett et al. (1985), a series of controlled experiments were undertaken worldwide for assessing the importance of the higher O_3 concentrations in eliciting a vegetation response. These controlled fumigation experimental results provided some of the evidence for emphasizing the importance of the higher concentrations in comparison to the mid- and low-level values (e.g., US EPA, 1986, 1992, 1996, 2013; Musselman et al., 1983, 1986, 1994; Hogsett et al., 1985; Nussbaum et al., 1995; Yun and Laurence, 1999; Lee and Hogsett, 1999; Oksanen and Holopaninen, 2001; Köllner and Krause, 2003; Wang et al., 2008).

These experiments help form the basis for the focus on the higher hourly average O_3 concentrations. In other words, by reducing the higher part of the distribution (not just the peak values), the risk to vegetation will be reduced based on the experimental evidence. Based on a thorough review of the literature, EPA (2013) concluded that (1) ozone effects in plants are cumulative; (2) higher ozone concentrations appear to be more important than lower

concentrations in eliciting a response; (3) plant sensitivity to ozone varies with time of day and plant development stage; and (4) quantifying exposure with indices that accumulate the ozone hourly concentrations and preferentially weight the higher concentrations improves the predictive power of exposure/response models for growth and yield, over using indices based on mean and other exposure indices. These conclusions have been reiterated in the current draft of the ISA (page 8-182). The draft ISA notes that no recent information available since the 2013 Ozone ISA alters these basic conclusions.

As indicated on page 8-183 in the draft ISA, the authors discuss the flux uptake metric. The metric is based on determining the O_3 concentration from the atmosphere that enters the leaf and is discussed in the draft ISA as follows:

> Another approach for improving risk assessment of vegetation response to ambient ozone is based on determining the ozone concentration from the atmosphere that enters the leaf (i.e., flux or deposition). Much work has been published in recent years, particularly in Europe, in using mathematically tractable flux models for ozone assessments at the regional, national, and European scale (Feng et al., 2017; Mills et al., 2011; Matyssek et al., 2008; Paoletti and Manning, 2007; Emberson et al., 2000b; Emberson et al., 2000a). While some efforts have been made in the U.S. to calculate ozone flux into leaves and canopies (Turnipseed et al., 2009; Uddling et al., 2009; Bergweiler et al., 2008; Hogg et al., 2007; Grulke et al., 2004; Grantz et al., 1997; Grantz et al., 1995), little information has been published relating these fluxes to effects on vegetation. Recently, Grantz et al. (2013) reported short-term ozone flux and related it to leaf injury in cotton in California. The authors reported that cotton leaves were most sensitive in the midafternoon, possibly due to changes in detoxification. They suggested with more research a sensitivity parameter may function well with the W126 metric. However, there remains much unknown about ozone stomatal uptake in vegetation at larger scales and how much uptake results in an injury or damage, which depends to some degree on the amount of internal detoxification occurring with each particular species. Those species having high amounts of detoxification potential may, in fact, show little relationship between ozone stomatal uptake and plant response (Musselman and Massman, 1999). The lack of data in the U.S. and the lack of understanding of detoxification processes have made this technique less viable for vulnerability and risk assessments in the U.S.

The interaction between O_3 and plant tissues is driven mainly by three distinct processes: changes in external O_3 concentration, O_3 uptake, and O_3 detoxification (see Heath et al., 2009). The diurnal pattern of detoxification does not necessarily match the diurnal patterns of external O_3 concentration and O_3 uptake (Heath et al., 2009; Wang et al., 2015; Dai et al., 2019). I would suggest that all three of these references be included in the draft ISA. In Lefohn et al. (2018), we discussed the stomatal flux index as follows:

> For assessing the potential for ozone to affect vegetation injury, growth and/or yield, exposure is defined as the integral of the instantaneous level over the period

> the vegetation is exposed to ozone (commonly expressed in unit of mol m^{-3} h or ppm-hrs) (Musselman et al., 2006). Examples of exposure indices are the W126 and AOT40 metrics (see Section 2.3.4). Although not necessarily considered exposure, seasonal average levels (e.g., 12-h daily average values averaged over a specified period) have also been referred to as exposure indices (US EPA, 2013). In contrast, the ozone dose is determined by first calculating the stomatal flux, which is a temporally dynamic measure of the rate of entry of ozone into the leaf (nmol m^{-2} s^{-1}). Dose is the total amount of ozone that is absorbed into the leaf through the stomata, in units of nmol m^{-2}, over a period of time and is calculated by integrating over time the instantaneous stomatal flux (Fowler and Cape, 1982; Mills et al., 2011b). The flux is accumulated over a species-specific phenological time window and the vegetation-damaging ozone flux is expressed as the Phytotoxic Ozone Dose (POD_Y), where Y represents a detoxification threshold below which it is assumed that any ozone molecule absorbed by the leaf will be detoxified (Mills et al., 2011b).

In Lefohn et al. (2018), we noted that flux-based metrics involve accumulation above a fixed flux threshold which is included to represent the detoxification capacity of the plant that varies with vegetation type/species (Mills et al., 2011b). While detoxification should ideally be represented as a dynamic variable rather than as a fixed threshold, modeling approaches are not yet able to take this dynamic variation into account for exposure-based (e.g., AOT40 or W126) or flux-based metrics. Results reported by Wang et al. (2015) for the diurnal changes of ascorbate, a major detoxification agent in the apoplast and leaf tissues of winter wheat, provide evidence for the dynamic nature of detoxification. Dai et al. (2019) have recently observed apoplastic ascorbate (ASC_{apo}) as an important contributor to the detoxification of O_3 in plants. The diurnal variation of ASC_{apo}, with maximum values occurring in the late morning with lower values experienced in the afternoon, was observed. With the detoxification potential by ASC_{apo} being lower in the afternoon, the implication is that the period of greatest uptake (e.g., the late morning/early afternoon hours) of O_3 in vegetation may coincide with the period of greatest detoxification potential, while the period of less detoxification may occur in the late afternoon hours, when the highest hourly O_3 concentrations occur. Heath et al. (2009) hypothesized that this was a possible explanation for the higher O_3 concentrations (which occurred in the later part of the day) eliciting a greater effect than the mid- and low-level values.

It is important to note that the US EPA made a conscious decision over 30 years ago, based on detailed analyses, to not use ***average*** concentration metrics as indicators of potential harm to vegetation. In 1986, the US EPA addressed the issue of using seasonal average concentration metrics to protect vegetation. In the early 1980s, the US EPA was considering the seasonal 7-h daily average concentration (referred to as the M7 (0900 h – 1559 h) metric) as a vegetation O_3 standard. In its Air Quality Criteria for Ozone and Other Photochemical Oxidants document (US EPA, 1986), the Agency stated on pages 6-10 and 6-11:

> A mean concentration (with various averaging times) is the most common statistic used. Because the mean is computed by summing the concentrations and dividing by time, it mathematically treats all concentrations as being equally effective in causing a plant response. The use of a mean concentration (with varying

averaging times) to characterize long-term exposures minimizes the contributions of peak concentrations to the response by treating low-level, long-term exposures the same as high-concentration, short-term exposures. ***The use of a longer-term mean concentration ignores the importance of peak concentrations and is inconsistent with the literature*** (emphasis added). A number of studies have shown that concentration is more important than exposure duration in causing a response. For example, studies with beans and tobacco (Heck et al., 1966) showed that a dose over a short time period induced more injury than the same dose distributed over a longer time period. Studies with tobacco showed that the O_3 concentration was substantially more important than exposure duration in determining the extent of foliar injury (Tonneijck, 1984). In this study, tobacco was exposed to a range of O_3 concentrations (0.02 to 0.15 ppm) for 8 hr/day for 1 to 7 days. In beans, foliar injury developed when the internal O_3 flux exceeded 115 ~moles/m^2 within 1 hr (Bennett, 1979). However, a single 3-hr exposure at about half the O_3 concentration (0.27 compared to 0.49 ppm) required approximately 64 percent greater internal O_3 flux to induce the same amount of foliar injury as in the 1-hr exposure (Bennett, 1979). Amiro et al. (1984) showed that higher concentrations were more important than low concentrations in causing injury. Their study also suggested the existence of a biochemical injury threshold (i.e., the O_3 uptake rates that plants can experience without inducing visible foliar injury). The greater importance of concentrations compared to exposure duration has been reported by other authors also (e.g., Heck and Tingey, 1971; Henderson and Reinert, 1979; Reinert and Nelson, 1979). The total ozone dose (concentration multiplied by time) has been used to describe plant exposure; however, it suffers from the same problem as the mean. The total dose is simply the summation of the ppm-hr over the study period, which treats all concentrations as being equally effective. Several investigators have attempted to give greater importance to peak O_3 concentrations. Oshima et al. (1977a,b) and Lefohn and Benedict (1982), for example, have summed only the ppm-hr of exposure greater than some preselected value. Larsen et al. (1983) introduced the concept of "Impact" to describe the effects of O_3 and SO_2 on soybeans. The "Impact (I)" is calculated similarly to total dose, except that the concentration is raised to an exponent greater than one ($I= C^W \times T$); this method of calculation effectively gives greater weight to the higher concentrations. More recently, Larsen and Heck (1984) have suggested the term "effective mean" as an approach for describing the greater importance of higher concentrations. The "effective mean" is defined as the average hourly impact raised to an exponent and divided by the duration. Severa1 lines of evidence suggest that higher concentrations have a greater influence in determining the impact of O_3 on vegetation. Studies have shown that plants can tolerate some combinations of exposure duration and concentration without exhibiting foliar injury or effects on growth or yield, illustrating that not all concentrations are equally effective in causing a response. From the toxicological perspective, it is the peaks or concentrations above some level that are most likely to have an impact. Effects occur on vegetation when the amount of pollutant that the plant has absorbed exceeds the ability of the organism to repair or compensate for the impact.

On page 6-12, the EPA (1986) continues

> Not only are concentration and time important but the dynamics of the O_3 exposure are also important; that is, whether the exposure is at a constant or variable concentration. Musselman et al. (1983) recently showed that fixed concentrations of O_3 cause the same kind of responses as variable concentrations at the equivalent dose. Fixed concentrations, however, had less effect on plant growth responses than variable concentrations at similar doses. Exposures of radishes to ambient O_3 in open-top exposure chambers showed that significant yield reductions occurred when the maximum O_3 concentration exceeded 0.06 ppm on at least 10 percent of the days when the crop was growing (Ashmore, 1984). Initial studies by Hogsett et al. (1985) have compared the response of alfalfa to daily peak and episodic O_3 exposure profiles which had the equivalent total O_3 dose over the growing season. Alfalfa yield was reduced to a greater extent in the episodic than the daily peak exposure. This study also illustrates the problem with the 7-hr seasonal mean concentration, which is that the peak concentrations are not properly considered. **The plants that displayed the greater growth reduction (in the episodic exposure) were exposed to a significantly lower 7-hr seasonal mean concentration** (emphasis added). Studies with SO_2 also showed that plants exposed to variable concentrations exhibited a greater plant response than those exposed to a constant concentration (Mclaughlin et al., 1979; Male et al., 1983). These results suggest that the mechanisms causing the response are the same, but that exposures to fixed concentrations underestimate the magnitude of plant growth responses that can occur with episodic exposures.

Since 1986, the US EPA has reiterated its commitment to focusing on weighting the higher hourly average concentrations more than the mid- and low-level hourly average concentrations to protect vegetation from both injury and damage (US EPA, 1996; 2006; 2013; Federal Register, 2015). As noted above, EPA (1986) noted the greater importance of concentrations compared to exposure duration. The total O_3 exposure (i.e., the sum of all hourly average concentrations over a time period), referred to as SUM00, in the literature, has been used to describe plant exposure. However, this exposure metric suffers from the same problem as the mean. The total exposure (SUM00) is simply the summation of the concentration multiplied by time over the study period, which treats all concentrations as being equally effective. Even though both the seasonal average (e.g., M7 and M12) and the SUM00 exposure metrics are still used in experimental studies for assessing vegetation risk, as EPA (1986, 1996, 2006, 2013) noted in its literature reviews, both O_3 exposure metrics are inappropriate for assessing vegetation effects based on biological experiments using different exposure regimes, as well as empirical “uncontrolled” experiments (e.g., the San Bernardino National Forest study).

It is not just the reduction of the "peaks," but also the reduction of those hourly average concentrations in the upper part of the distribution that is important to reduce vegetation effects. To accomplish this goal, the Agency moved from its initial consideration of seasonal M7 (daily 7-h average concentration averaged over a growth season) or M12 (daily 12-h average

concentration averaged over a growth season) exposure metrics to the use of cumulative exposure metrics (i.e., SUM06 and W126). The SUM06 exposure metric is the sum of all hourly average concentrations greater than or equal to 60 ppb. As mentioned above, the US EPA has discussed the use of flux-based indices in its standard-setting process but believes that further research concerning dynamic detoxification and other considerations are required before flux indices might be considered as a practical use in the standard-setting process. The W126 O_3 exposure index, described in Lefohn and Runeckles (1987) and Lefohn et al. (1988), is a weighted cumulative exposure index that provides greater weight to the higher hourly average O_3 concentrations rather than the mid- and lower-level values. Figure 2-1 below illustrates the weighting scheme. In addition, the W126 index does not impose an artificial threshold and is not an "average" of several values collected over the course of a short- or long-term time period. The W126 weighting scheme, as noted by the EPA (2013), is supported by research results performed under ***controlled*** conditions, as well as under ***uncontrolled*** exposure conditions, such as observed in the San Bernardino National Forest in the Los Angeles area.

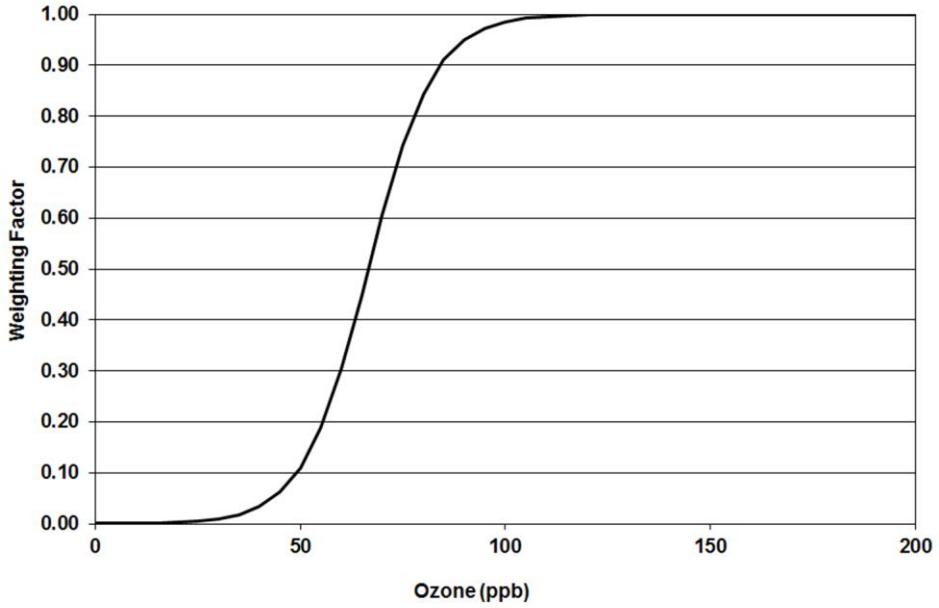


Figure 2-1. The weighting applied to hourly average ozone values for the calculation of the W126 exposure index (see Lefohn et al., 1988 for further details).

The 2013 ISA noted that at the San Bernardino site, located near Los Angeles, reductions in ambient O_3 exposures between 1980 and 2000 were related to improvements in tree conditions. The frequency of midrange hourly average ozone concentrations was little changed over this period. EPA (2013) suggested it was the reduction in the higher hourly average ozone concentrations that was responsible for the improvement in tree health. Please note that the reference to the empirical results at the San Bernardino National Forest were not included in the current draft of the ISA**. I suggest that t*he San Bernardino National Forest results should be discussed in the draft of the ISA as providing independent confirmation separate from the chamber experimental studies of the importance of the higher hourly average O_3 concentrations in influencing vegetation effects***.

As indicated above, based on a thorough review of the vegetation literature, (1) ozone effects in plants are cumulative; (2) higher ozone concentrations are more important than lower concentrations in eliciting a response; (3) plant sensitivity to ozone varies with time of day and plant development stage; and (4) quantifying exposure with indices that accumulate the ozone hourly concentrations and preferentially weight the higher concentrations improves the predictive power of exposure/response models for growth and yield, over using indices based on mean and other exposure indices. Based on the peer-reviewed literature and its own research studies, the EPA (2013) identified the W126 cumulative exposure metric as the most appropriate to use to evaluate both the adequacy of the current secondary standard and the appropriateness of any potential revisions (Federal Register, 2015 – page 65373). In its current O_3 NAAQS review, both the draft ISA and draft PA continue to focus on the W126 cumulative exposure index as the metric to protect vegetation.

2.3 Air Quality Index (AQI) focuses on the higher ozone concentrations

EPA's recognition of the importance of the higher O_3 concentrations is reflected its Air Quality Index (AQI) reporting across the US (EPA, 2018). Local air quality agencies are required to report air quality using the Air Quality Index (AQI) as required in 40 CFR Part 58.50 and according to 40 CFR Appendix G to Part 58. Metropolitan Statistical Areas (MSAs) with a population of more than 350,000 are required to report the AQI daily to the general public. MSAs must report the AQI daily, which is defined as at least five days each week. There are six AQI categories and their names and colors are as follows:

AQI Range	**Descriptor**	**Color**
0 to 50	Good	Green
51 to 100	Moderate	Yellow
101 to 150	Unhealthy for Sensitive Groups	Orange
151 to 200	Unhealthy	Red
201 to 300	Very Unhealthy	Purple
301 to 500	Hazardous	Maroon

The pollutant specific sensitive groups are separated by 8-h daily maximum O_3 concentrations as indicated in Fig. 2-2 below.

8-hour Ozone Concentration	Air Quality Index Levels
0 – 54 ppb	Good (Green)
55 – 70 ppb	Moderate (Yellow)
71 – 85 ppb	Unhealthy for Sensitive Groups (Orange)
86 – 105 ppb	Unhealthy (Red)
106 – 200 ppb	Very Unhealthy (Purple)
>200 ppb	Hazardous (Maroon)

Figure 2-2. Air quality index levels (AQI) related to 8-hour concentrations.

An important aspect of the AQI index is that the higher the 8-h daily maximum O_3 concentration the higher the index. While this would appear to be an obvious statement, the ramification is that simply counting the number of exceedances of 8-h daily maximum concentrations greater than 70 ppb will not provide an accurate indication of the health risks associated with O_3 exposures during a specific period (e.g., March-October). ***Rather, it is the cumulative sum of the number of days above 70 ppb weighted by a factor that relates to each day's index range that is most important.*** For example, in Fig. 2-3 below for Los Angeles-Long Beach-Anaheim, CA (EPA, 2019c), when one compares the exposure for 2015 with the exposure for 2017, one might conclude based on the number of exceedance days (108 versus 107 above 70 ppb) that the annual O_3 exposures were similar. However, more days in 2017 occurred with orange and purple exceedances than in 2015. Based on the number of days that experienced orange and purple exceedances, the O_3 exposures (i.e., health risk) experienced by the public were higher in 2017 than 2015. As indicated, simply comparing the number of exceedance days among O_3 monitors is not an adequate way to quantify the exposure differences among monitors. In an effort to inform the public of potential health risks, the American Lung Association's annual State of the Air Report (ALA, 2019) (http://www.lung.org/our-initiatives/healthy-air/sota/key-findings/) utilizes weighting factors applied to each range of daily 8-h daily maximum O_3 concentrations associated with the Air Quality Index levels described above. The number of orange days (unhealthy for sensitive groups) experienced by each county receives a factor of 1; red days (unhealthy), a factor of 1.5; purple days (very unhealthy), a factor of 2; and maroon days (hazardous), a factor of 2.5. This weighting scheme provides a better way to inform the public of potential health risks than the simple determination by various groups to count the number of exceedances above 70 ppb.

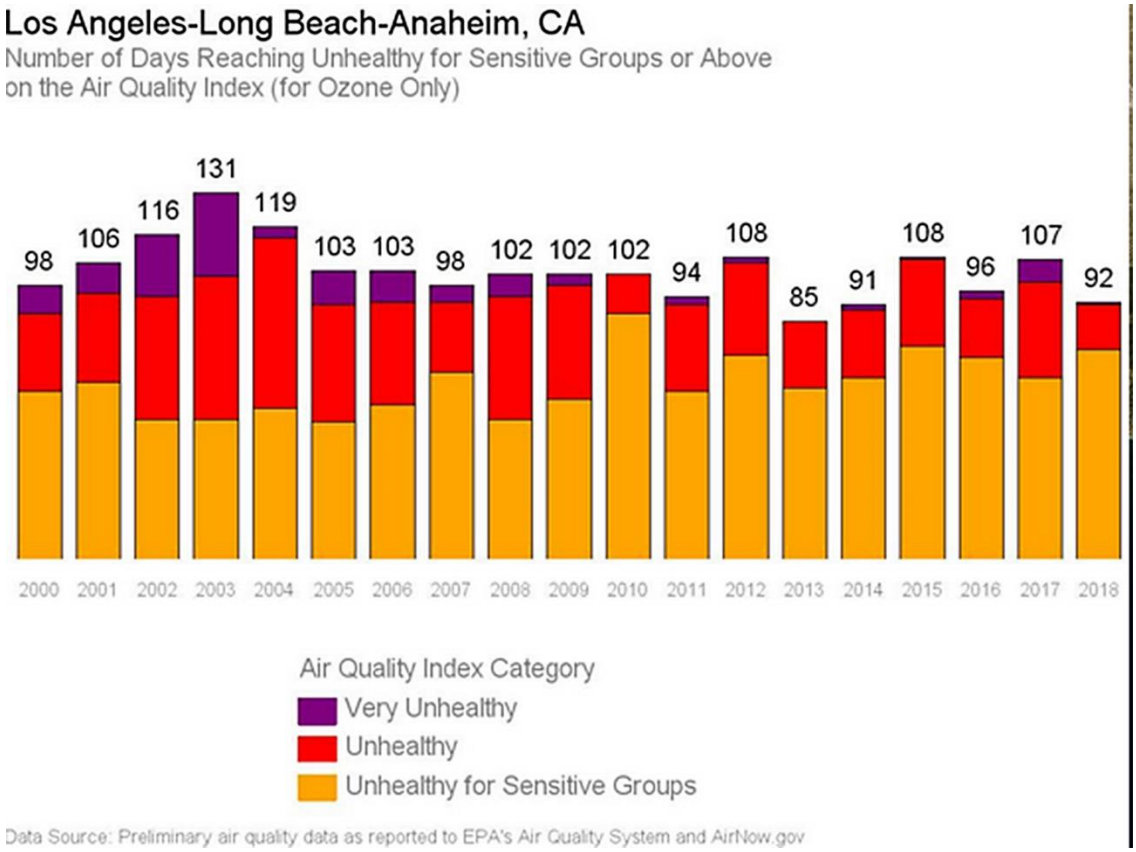


Figure 2-3. Number of days reaching unhealthy levels for sensitive groups or above on the Air Quality Index for Los Angeles-Long Beach -Anaheim, CA. Source: U.S. EPA, 2019: A Look Back: Ozone in 2018.
https://epa.maps.arcgis.com/apps/Cascade/index.html?appid=9bec4031ba6f4887a9f332a8f058b198

2.4 Further Ramifications for the Importance of the Higher Concentrations

As noted above, both vegetation effects research and controlled laboratory studies of human volunteers indicate that higher O_3 hourly average concentrations elicit a greater effect on hour-by-hour physiologic response than lower hourly average values. The weighting of the higher values compared to the mid and lower hourly average O_3 concentrations results in a nonlinear response for both human health and vegetation (Hazucha and Lefohn, 2007; Lefohn, Hazucha, Shadwick, and Adams, 2010; Heath et al., 2009). The nonlinear response noted above for the human health clinical studies and the vegetation experiments has an important impact on the validity of Haber's rule (also referred to as Haber's law). Haber's rule states that, for a given poisonous gas, $C \times t = k$, where C is the concentration of the gas (mass per unit volume), t is the amount of time necessary in order to produce a given toxic effect, and k is a constant, depending on both the gas and the effect. Haber's law or rule, as commonly understood in inhalation toxicology, states: C×T=constant, meaning that identical products of concentration of an agent in air (C) and duration of exposure (T), the CT product, will yield an identical biological response. The formula was originally developed by the German physical chemist Fritz Haber (1868 –1934)

to characterize the acute toxicity of chemicals used in gas warfare. For example, the rule states that doubling the concentration will halve the time. Haber's rule is an approximation and Haber himself acknowledged that it was not always applicable (https://en.wikipedia.org/wiki/Haber%27s_rule). The greater importance of concentrations compared to exposure duration results in the failure of Haber's rule. Specifically, when concentration is more important than the time required to elicit an adverse effect, Haber's rule will not be applicable when attempting to determine a cumulative exposure. Miller et al. (2000) discuss the fact that many toxicologists have used Haber's rule to analyze experimental data whether their chemicals, biological endpoints, and exposure scenarios were suitable candidates for the rule. As indicated by the literature, as well as the EPA reviews since 1986, Haber's rule does not appear to be applicable for O_3.

Both the vegetation and the clinical health studies show that because the higher hourly average O_3 concentrations have a greater effect than the mid- and lower-level values, the use of *long-term average concentrations, which combine all hourly values into one number, is an inappropriate index* to use because the highest hourly average concentrations, which are more biological important than the lower values, are "averaged" out by smoothing, mathematical effects. The important result is that a comparison among different O_3 monitoring sites results in many of the sites having similar long-term average concentrations that differ in the magnitude and number of the biologically important elevated O_3 hourly average concentrations. The long-term average is not correlated with the number and magnitude of the biologically important elevated hourly average concentrations.

Following the setting of the 2015 O_3 standard, the metrics used in the United States to assess the risk of O_3 to human health and vegetation continue to be the 8-h daily maximum concentration (human health) and the W126 cumulative exposure index (vegetation). Both metrics as discussed above are biologically relevant. However, other exposure metrics are used for assessments by researchers (e.g., see TOAR paper by Lefohn et al., 2018; Wang et al., 2019; Lefohn et al., 2017). Using the same hourly data, it is possible to reach entirely different scientific conclusions for assessing trends and O_3 impacts utilizing different exposure metrics (Lefohn et al., 2017, 2018). ***What is most important is that the exposure metrics be biologically relevant and defensible***.

The impacts of surface O_3 on human health and vegetation have prompted precursor emission reductions in the United States. As emissions change, the distribution of hourly O_3 concentrations also changes, as do the values of individual exposure metrics (Lefohn et al., 2017). The distribution changes can result in exposure metric trend patterns changing in a similar direction as trends in emissions (e.g., metrics increase as emissions increase) or, in some cases, in *opposite* directions. For example, Lefohn et al. (2017) reported, using the current form of the 8-h standard for the 196 US sites studied in their analysis, 162 sites showed negative trends, 32 showed no trends, and 2 had insufficient data. In comparison, using the a 6-month (April-September) 12-h daylight average concentration, 92 sites showed negative trends, 85 sites showed no trend, 19 sites showed increasing trends, and 0 sites showed insufficient data. Thus, while 162 sites exhibited a negative trend in O_3 using the 4th highest annual 8-h average exposure metric, using the 6-month 12-h daylight average exposure metric based on data from the same sites showed only 92 instances of negative trends and increasing trends for 19 sites compared to

0 sites for the 8-h form of the standard. The trend patterns of the two exposure metrics to changes in the reduction of emissions using the same data from the US sites were extremely different and provided an excellent example why the selection of appropriate biologically relevant exposure metrics is important for assessing human health and/or vegetation effects.

In addition to the Lefohn et al. (2017) analysis, Lefohn et al. (2018) compared several exposure metrics that focused on the higher hourly average O_3 concentrations with two metrics that are based on all hourly average concentrations (i.e., mean and median). In Table 5 of their analyses, Lefohn et al. (2018) compared the trends using different metrics that were in the same direction (i.e., decreasing, increasing, or no significant change) compared to other metrics. Trends in the human health metric impacted by the high end of the distribution bear the least resemblance to trends in the mean and median values with generally less than 50% of sites having trends in the same direction. In many cases the trends were in opposite direction (i.e., the metrics associated with the highest concentrations exhibited decreases over time, while the mean and median metrics exhibited increases). Table 1-1 below provides a summary of the relationship between two of the exposure metrics focused on the highest concentrations (i.e., the annual 4th highest daily maximum 8-h average concentration (4th dma8epa) and the number of daily maximum 8-h averages greater than 70 ppb (nvgt070 summer) for the months of April-September) and the median and mean values. As indicated above, overall, trends in the four mean/median metrics were not representative of the trends behavior of those metrics associated with the higher concentrations.

Table 1-1. Comparison of trends using different metrics that were in the same direction (i.e., decreasing, increasing, or no significant change) compared to other metrics. Source: Adopted from Lefohn et al. (2018).

	median annual	mean annual	median summer	mean summer
4th dma8epa	33%	39%	43%	50%
nvgt070 summer	37%	44%	43%	53%

Lefohn et al. (2017) cautioned that trends in mean or median concentrations did not appear to be well associated with those exposure metrics that are most optimum indicators of overall changes in anthropogenic emissions, biological effects, or climate-driven meteorology. Similar to the findings of other studies, Lu et al. (2018) reported that exposure indices, such as the median and the M12 average metrics (average of hourly O_3 concentrations for the 12-h period from 08:00 to 19:59 local time April−September), which focus on the midrange of the O_3 hourly average concentration distribution, did not appear to adequately describe the magnitude and frequency of high O_3 events. The authors reported that the median and seasonal M12 metrics, instead of being much greater in China than in Japan, South Korea, Europe, and the United States, were similar in values for all countries, whereas much higher hourly averaged O_3 concentrations were experienced in China compared to the other four countries.

Both the human health clinical results and the vegetation experiments form the basis for the implementation of O_3 control strategies in the United States and around the world. Simply stated, by reducing the higher part of the distribution (not just the peak values), it is believed that the risk to human health and vegetation will be reduced. As the number of repeated peaks is reduced, the risk to human health and vegetation is reduced. In 2015, it was the opinion of the EPA (Federal Register, 2015 – page 65358) that both acute and chronic effects would be reduced in implementing the new O_3 standards. The higher part of the distribution of hourly average concentrations would be moved downwards toward the middle hourly average concentrations. While some may not agree with the EPA and that to eliminate "chronic" effects from O_3 one must reduce the lower hourly average concentration. However, this may not be achievable. By reducing NO_x to reduce the higher O_3 concentrations, there is a reduction in NO_x scavenging and a resultant upward shift of the lower concentrations. If one believes that an annual average or a seasonal average of the hourly average concentrations are the only candidates for exposure metrics to use for controlling chronic effects, then models, as well as empirical data, indicate that these metrics will not perform as some researchers anticipate. However, if one believes, as the EPA believed in 2015, that adequate protection for both acute and chronic exposures is attained by protecting against repeated occurrences of exposures of concern (i.e., the higher hourly average concentrations), then practical emission control strategies will reduce both acute and chronic effects.

3 Fundamental Principle No. 2: Daily maximum hourly averaged ozone concentrations will remain well above 0 parts per billion (ppb) even if all anthropogenic emissions were eliminated worldwide

3.1 Introduction

As noted above, the ramifications of Fundamental Principle No. 2 are important. If one assumes that to control for chronic health effects that annual or seasonal averages must be reduced, then models, as well as empirical data, indicate that annual and seasonal average metrics will not perform as some researchers anticipate. Lefohn et al. (2017) noted for 196 O_3 monitors in the US that trend patterns were more variable using the 12-h seasonal mean exposure metric than the annual 4th highest 8-h daily maximum concentration (which focuses on the highest concentrations). While there were no sites that experienced increasing trends in the 4th highest 8-h daily maximum metric, 10% of the 196 sites in the US experienced increasing trends using the 12-h seasonal mean index. There were also more sites that experienced no trends using the 12-h seasonal mean metric than the 4th highest 8-h daily maximum index. In other words, the metric that focuses on the highest concentrations behaved more consistently than the metric that focused on the entire range of concentrations. As more and more anthropogenic emissions are reduced, there is a range of hourly average O_3 concentrations at a specific monitoring site influenced by these emissions that begins to appear resistant to further change. As will be discussed below, the process of identifying this range of O_3 concentrations begins with the movement of both the high and lower hourly average concentrations towards the mid-level values as emissions are reduced. These patterns were reported in the literature by various

researchers (e.g., Lefohn et al., 1998; EPA, 2014b; Simon et al., 2015; Lefohn et al., 2017, 2018).

To develop the discussion for Fundamental Principle No. 2, we first explore the changing patterns in the distribution of hourly average O_3 concentrations as emissions are reduced. We explore the changes in the frequency of both high and low levels of O_3 hourly average concentrations. Following the description of the changes in the distribution patterns, we then explore when during the year the highest hourly average O_3 concentrations occur at sites where emissions were reduced. Both modeling and empirical data are presented for comparison. Finally, we explore what the results from models, as well as empirical data, tell us about background O_3 levels. Integrating all the information described in this section provides us with the modeling and empirical support for Fundamental Principle No. 2: **Daily maximum hourly averaged ozone concentrations will remain well above 0 parts per billion (ppb) even if all anthropogenic emissions were eliminated worldwide**.

The importance of Fundamental Principle No. 2 is that the continuation of emissions reductions will fail in achieving reductions of the lowest hourly average O_3 concentrations. Emission reductions will achieve the shifting of the higher hourly average O_3 concentrations toward the mid-level values. However, rather than emission reductions causing the mid-level hourly values to shift downward toward the lower values, the lower hourly average concentrations will shift upwards toward the mid-level values, with the result that a Gaussian-like (i.e., bell shaped) distribution of hourly average concentrations may occur depending upon the amount of emission reductions and the influence of the remaining anthropogenic contributions to ambient O_3 levels. This phenomenon will be discussed in my comments on the draft PA in the coming weeks. The distribution of background O_3 hourly average concentrations will determine for each site the range of hourly average concentration values. The shape of the distribution of O_3 hourly average concentrations may appear to be Gaussian-like with only the amplitude varying at each site. Thus, daily maximum hourly averaged O_3 concentrations will remain well above 0 ppb even if all anthropogenic emissions were eliminated everywhere. Because we never will be able to reduce all anthropogenic emissions (either in the US or outside of the US), we can explore how O_3 distributions of hourly average concentrations have changed as emissions have been reduced. Models, as well as empirical data, provide us insight about the behavior of these changes.

3.1 Patterns of the Shifting of Hourly Average Concentrations as Emissions are Reduced

3.1.1 The Lower Ozone Concentrations Shift Upward as Emissions are Reduced

In the 2014 EPA Policy Assessment document (EPA, 2014a), the EPA noted in its modeling effort that as NO_x was reduced, the high end of the distribution shifted downward and the low-end of the distribution shifted upward. There was a compression of the distribution of concentrations. Figures 4-9 and 4-10 (pages 4-24 and 4-25) presented in EPA (2014b) are reproduced here as Figs. 3-1 and 3-2. For the 12 urban-influenced sites described in the modeling results for the period April-October, the general pattern from the modeling effort is that as

emissions reductions occur to attain 4th highest 8-h daily maximum (MDA8) values for alternative scenarios of 75, 70, 65, and 60 ppb, the individual daily MDA8 values at the high end of the distribution are reduced toward the center of the distribution. However, the lowest daily MDA8 values increase. The model predictions have been observed using actual observations resulting from emission reductions (Lefohn et al., 1998; Simon, 2015; Lefohn et al., 2017, 2018). The shifting of the lower concentrations toward the mid-level values is associated with less NO_x scavenging of the lower hourly average concentrations as reduction in NO_x emissions occurs (Lefohn et al., 1998; U.S. EPA, 2014b; Simon, 2015; Lefohn et al., 2017, 2018).

By focusing on a specific level of the 4th highest MDA8 value that protects human health, the US EPA's emissions reductions strategy forces the concentrations of concern (i.e., the highest values) downward toward the middle values. The frequency of the middle concentration values is increasing due to the downward shift from the higher values as indicated above, as well as the upward shift of the lower MDA8 concentrations toward the middle. In other words, the high end is coming down and the low end is coming up. Both meet in the middle of the distribution as described by Lefohn et al. (1998) and Simon et al. (2015).

On page 3C-98 (3C.7.2 Distribution of Hourly O_3 Concentrations) of the draft PA (EPA, 2019), the Agency has updated its 2014 modeling analyses. Its current conclusions are similar to the conclusions reached in the 2014 PA (EPA, 2014a) earlier document. Figures 3C-67 (page 3C-100) through Figure 3C-74 (page 3C-107) display diurnal boxplots of hourly O_3 concentrations for 2015-2017 at monitor locations in each urban area. For each hour of the day, the rectangular box represents the 25th and 75th percentiles of the distribution, with a solid line representing the median of the distribution through the center. Each box has "whiskers" which extend up to 1.5 times the interquartile range (i.e., the 75th percentile minus the 25th percentile) from the box, and dots which represent outlier values. Black boxplots represent observed hourly O_3 concentrations, while blue boxplots represent hourly O_3 concentrations adjusted to meet the current standard of 70 ppb. Red boxplots represent hourly O_3 concentrations adjusted for the 75 ppb scenario, and green boxplots represent hourly O_3 concentrations adjusted for the 65 ppb scenario.

Eight cities were highlighted in the Agency's modeling analyses (Atlanta, Boston, Dallas, Detroit, Philadelphia, Phoenix, Sacramento, and St. Louis). Ambient hourly O_3 monitoring data for years 2015 through 2017 in each of the eight urban study areas was adjusted using a model-based adjustment approach to create three different air quality scenarios. These scenarios included conditions that just meet the current O_3 standard (design value of 70 ppb), as well as conditions that just meet two alternative standards (design values of 75 ppb and 65 ppb). The figures below illustrate in the draft Policy Assessment document (EPA, 2019b) the shifting of the hourly average concentrations that result in the compression of the distribution as emissions are reduced to attain the three scenarios listed above. The EPA described the compression of the distribution of hourly average concentrations (high-end shifting downward and the low-end shifting toward the middle) in the modeling results in the draft PA (page 3C-98) as follows:

> The hourly plots show similar patterns in most of the urban areas. O_3 concentrations during daytime hours decrease from observed values (black) to values adjusted to meet the current standard of 70 ppb (blue)

and decrease further under the alternative scenario of 65 ppb (green). These daytime decreases are mainly seen on high O_3 days represented by outlier dots extending above the box and whiskers. Some areas had observed 2015-2017 design values already meeting the alternative scenario of 75 ppb, therefore some plots show increases in O_3 concentrations while other areas show decreases in O_3 concentrations for the 75 ppb scenario.

In some urban areas O_3 concentrations on the mid-range days, represented by the 25th –75th percentile boxes, remained fairly constant (e.g. Boston) while in other urban areas O_3 on mid-range days decreased (e.g. Atlanta). Although daytime O_3 decreased, concentrations during morning rush-hour period generally increase. These increases are associated with VOC-limited and NO_X titration conditions near NO_X sources during rush-hour periods. Reducing NO_X under these conditions results in less O_3 titration and thus increases O_3 concentrations. Nighttime increases in O_3 as a results of NO_X reductions are often seen to a lesser extent than morning rush hour period increases. Collectively these features generally lead to a flattening of the diurnal O_3 pattern with smaller differences between daytime and nighttime concentrations as NO_X emissions are reduced. Urban areas that required more substantial NO_X reductions for the 65 ppb scenario generally had more pronounced patterns of decreases in daytime O_3 and increases in nighttime O_3 leading to a flatter diurnal O_3 pattern (e.g. Sacramento in Figure 3C-73).

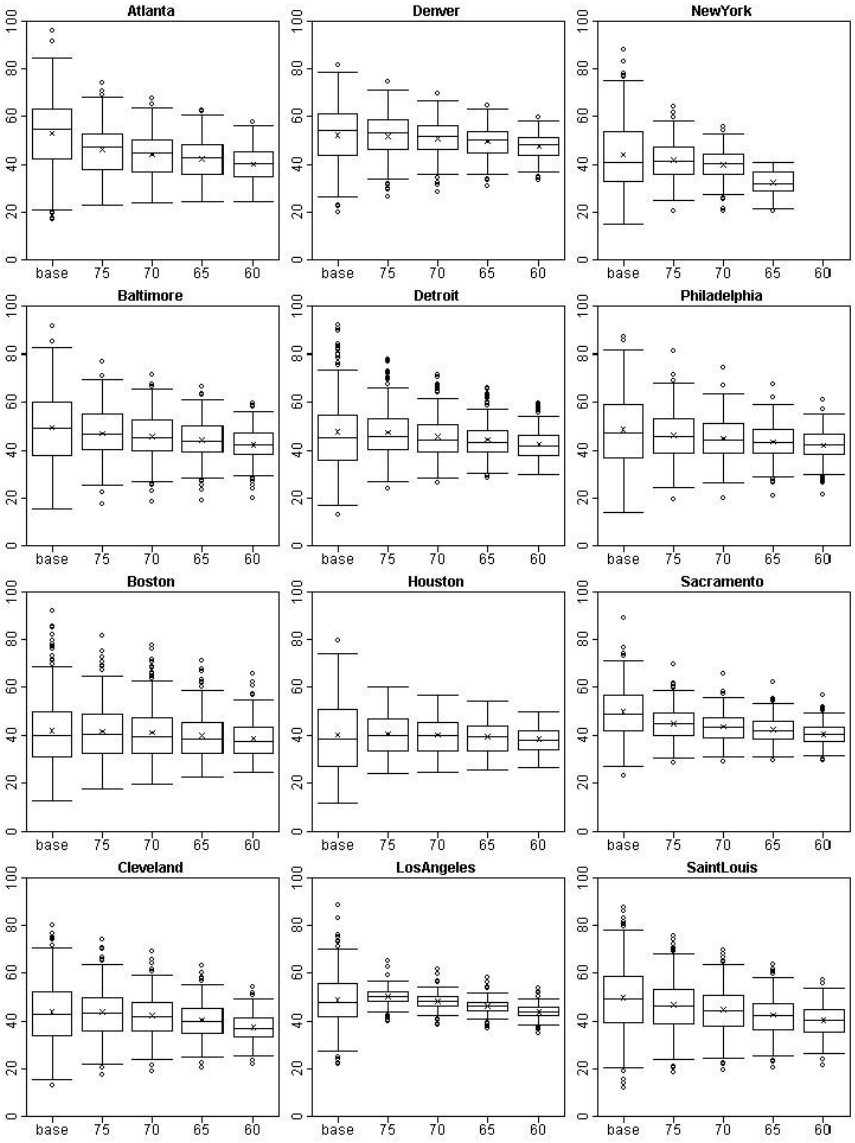


Figure 4-9. Distributions of Composite Monitor Daily Maximum 8-hr Average Values for the 12 Urban Study Areas in the Epidemiology-based Risk Assessment. Plots depict values based on ambient measurements (base), and values obtained with the HDDM adjustment methodology when just meeting the 75, 70, 65 and 60 ppb standards. Values shown are based on CBSAs for April-October of 2007. Note that the HDDM adjustment technique was not able to adjust air quality to just meet a 60 ppb standard in New York, so no boxplot is shown for that case. Boxes represent the median and quartiles, X's represent mean values, whiskers extend up to 1.5x the inter-quartile range from the boxes, and circles represent outliers.

Figure 3-1. Figure 4-9 from US EPA (2014b).

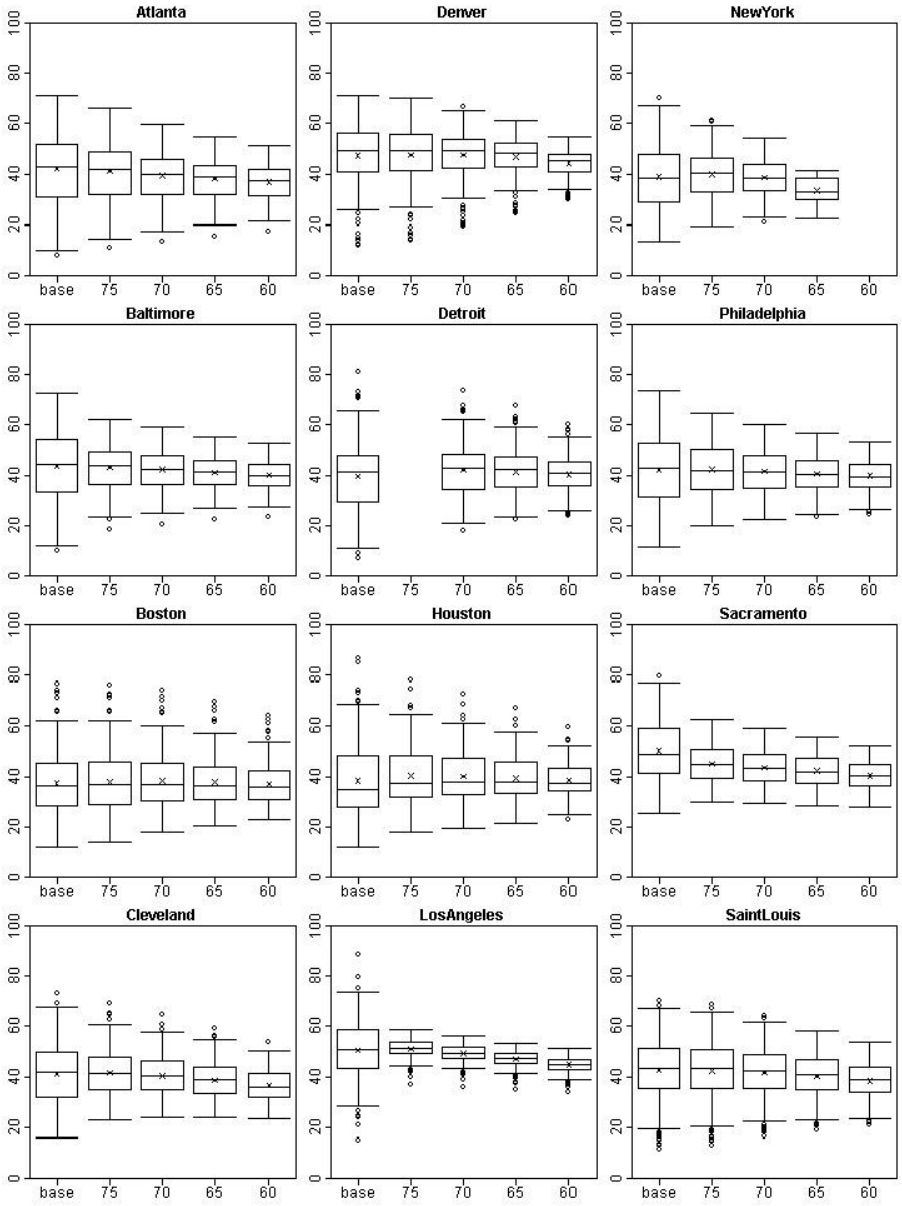


Figure 4-10. Distributions of Composite Monitor Daily Maximum 8-hr Average Values for the 12 Urban Study Areas in the Epidemiology-based Risk Assessment. Plots depict values based on ambient measurements (base), and values obtained with the HDDM adjustment methodology when just meeting the 75, 70, 65 and 60 ppb standards. Values shown are based on CBSAs for April-October of 2009. Note that Detroit air quality was meeting 75 ppb in 2008-2010, and the HDDM adjustment technique was not able to adjust air quality to just meet a 60 ppb standard in New York, so no boxplots are shown for those cases. Boxes represent the median and quartiles, X's represent mean values, whiskers extend up to 1.5x the inter-quartile range from the boxes, and circles represent outliers.

Figure 3-2. Figure 4-10 from US EPA (2014b).

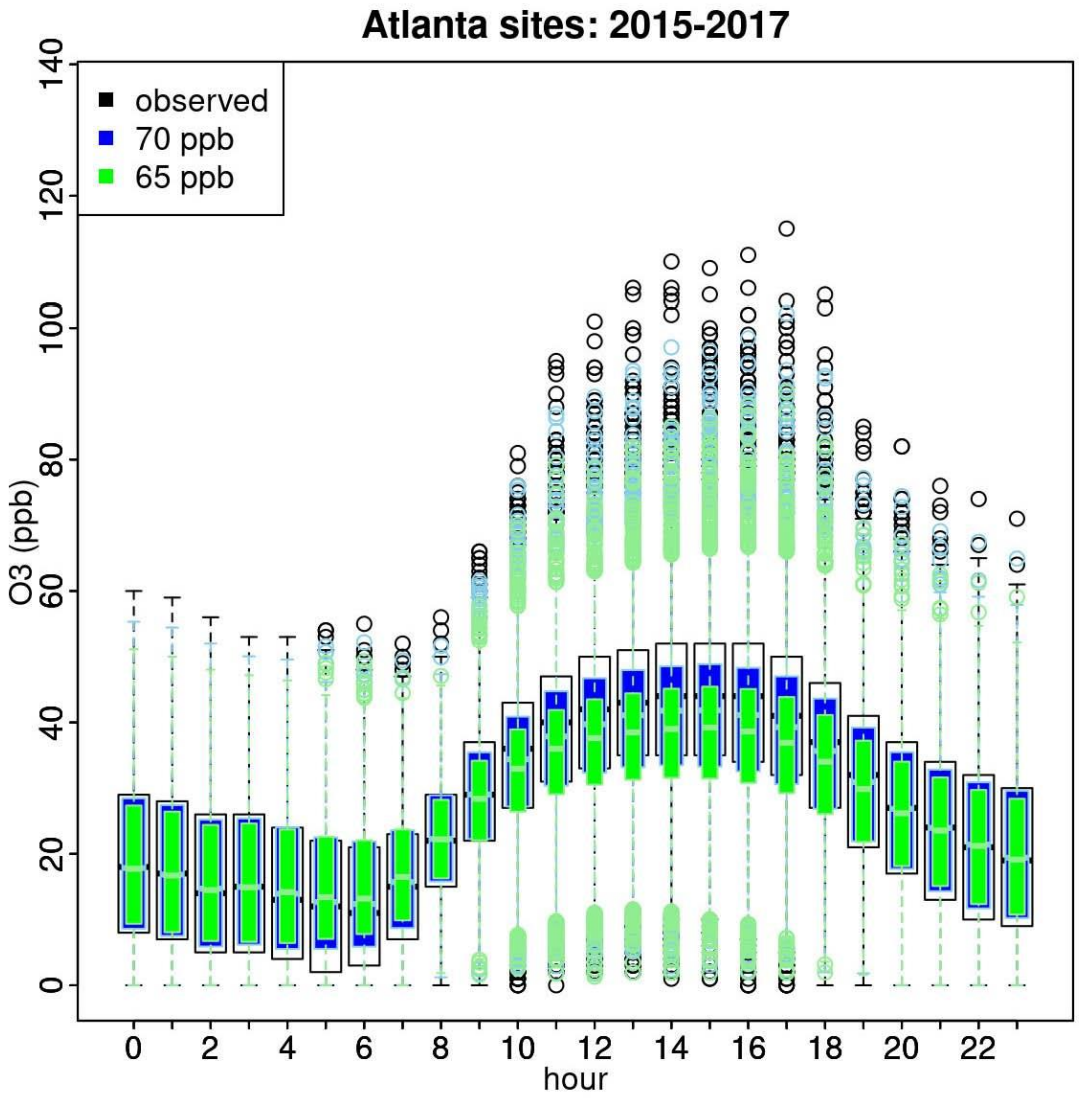


Figure 3C-67. Diurnal distribution of hourly O_3 concentrations at monitoring sites in Atlanta.

Figure 3-3. Figure 3C-67 from US EPA (2019b).

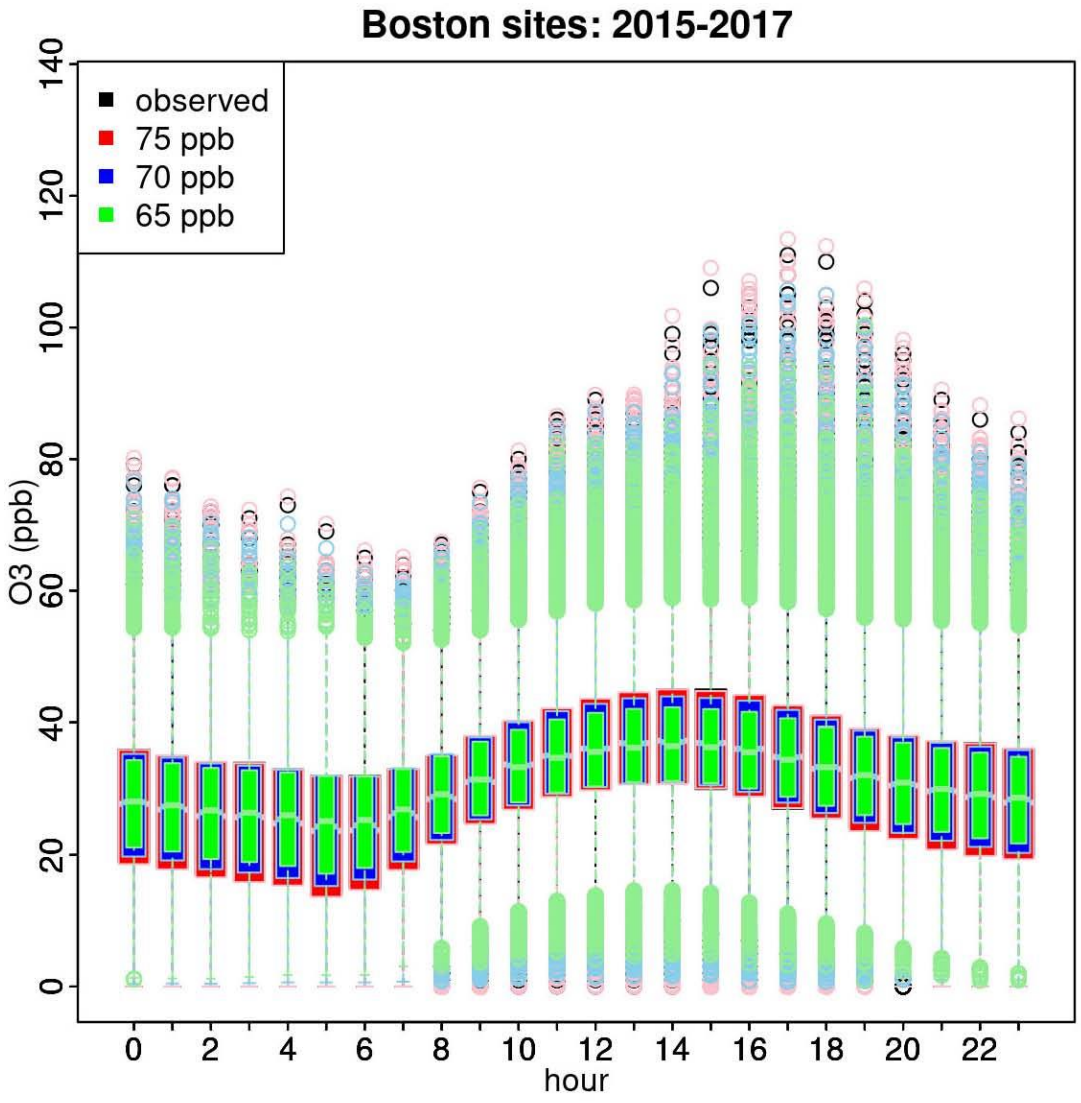


Figure 3C-68. Diurnal distribution of hourly O_3 concentrations at monitoring sites in Boston.

Figure 3-4. Figure 3C-68 from US EPA (2019b).

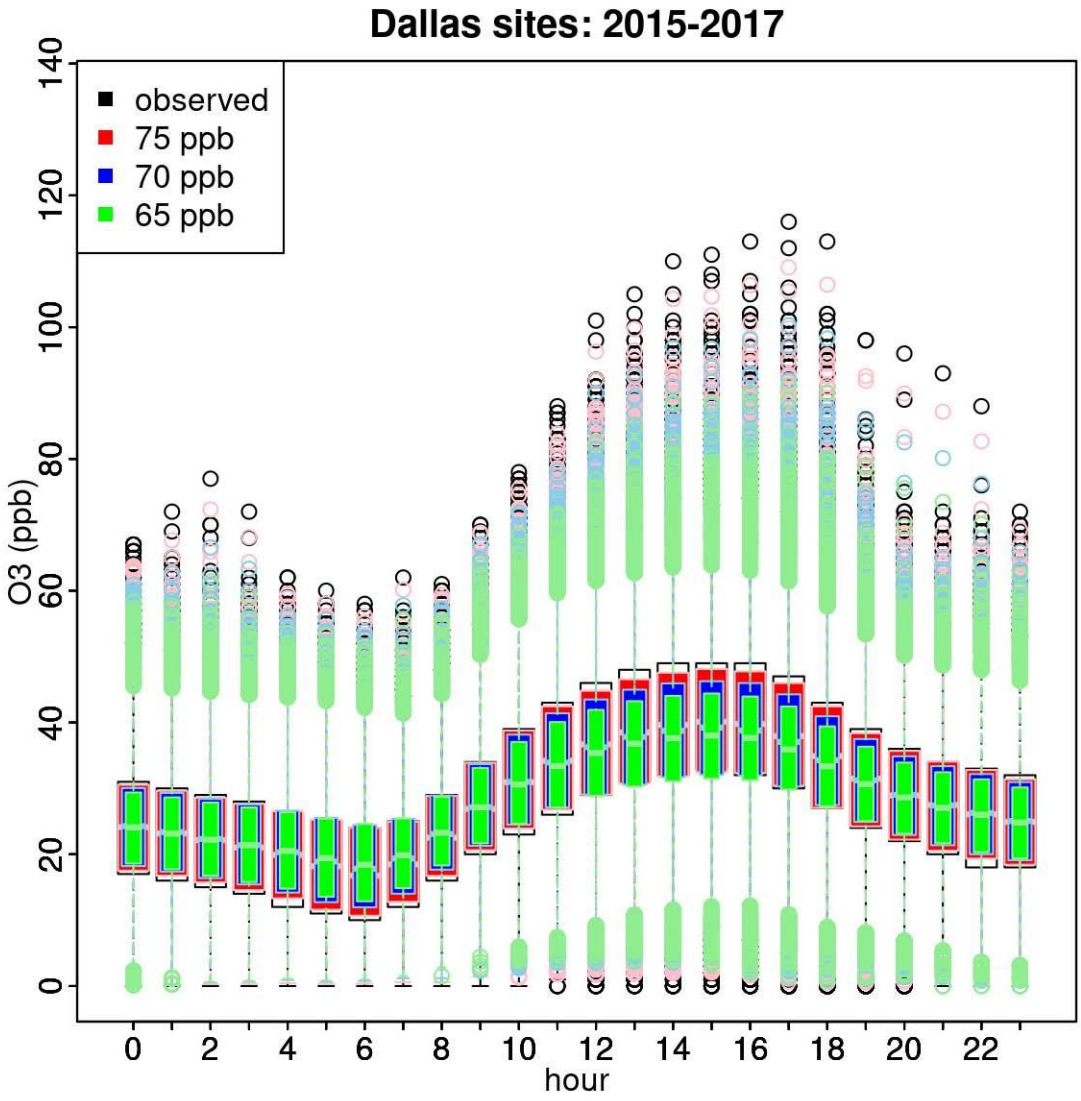


Figure 3C-69. Diurnal distribution of hourly O_3 concentrations at monitoring sites in Dallas.

Figure 3-5. Figure 3C-69 from US EPA (2019b).

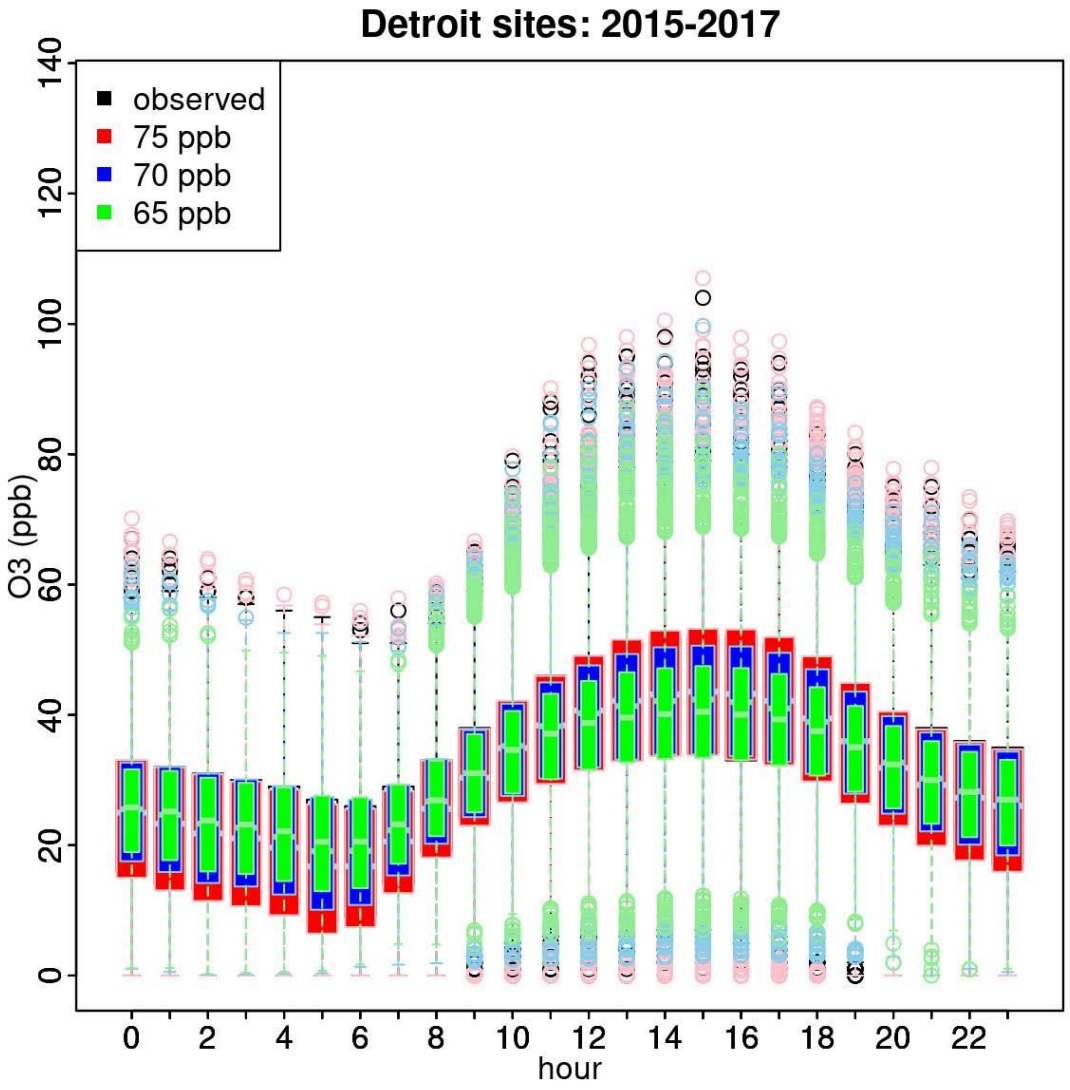


Figure 3C-70. Diurnal distribution of hourly O_3 concentrations at monitoring sites in Detroit.

Figure 3-6. Figure 3C-70 from US EPA (2019b).

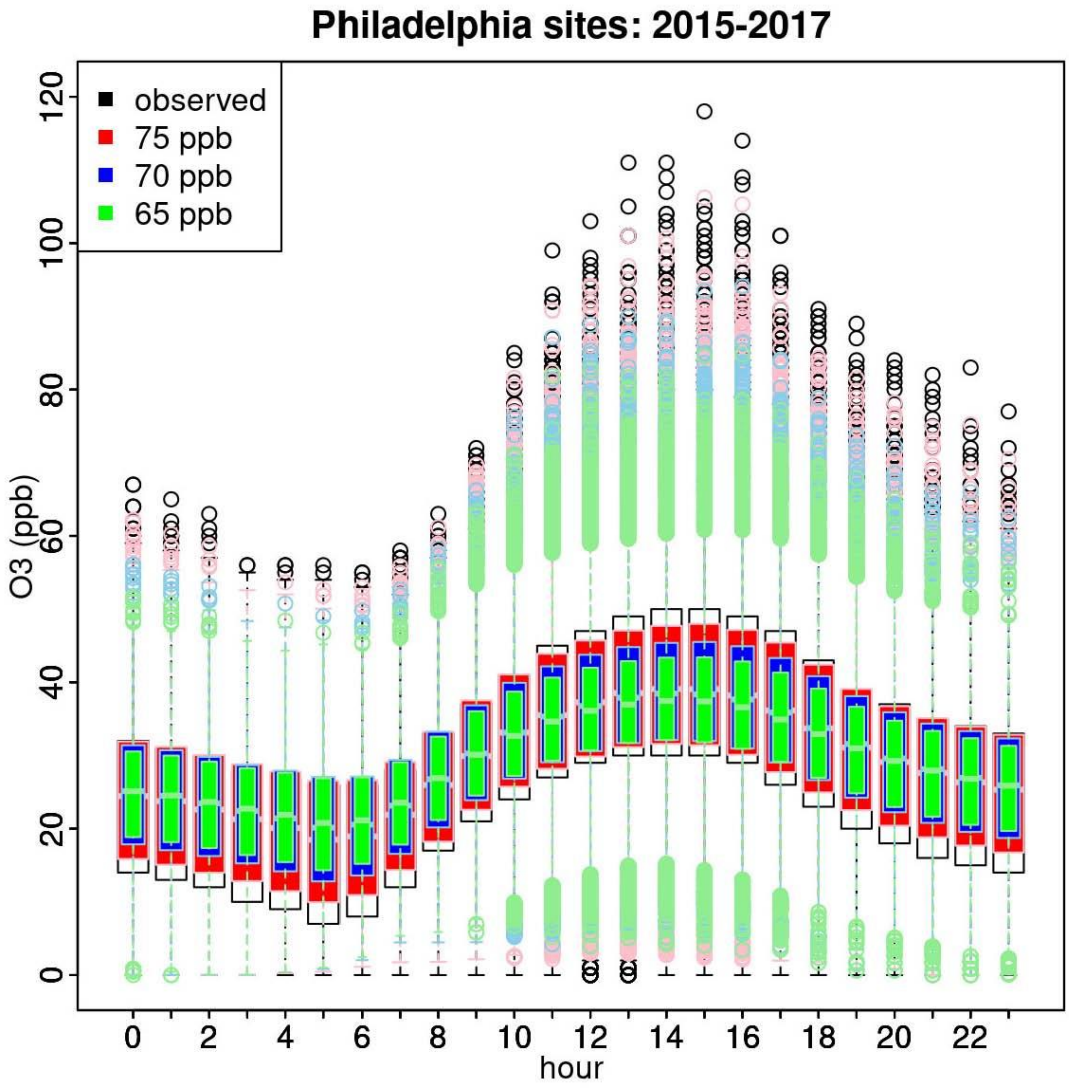


Figure 3C-71. Diurnal distribution of hourly O_3 concentrations at monitoring sites in Philadelphia.

Figure 3-7. Figure 3C-71 from US EPA (2019b).

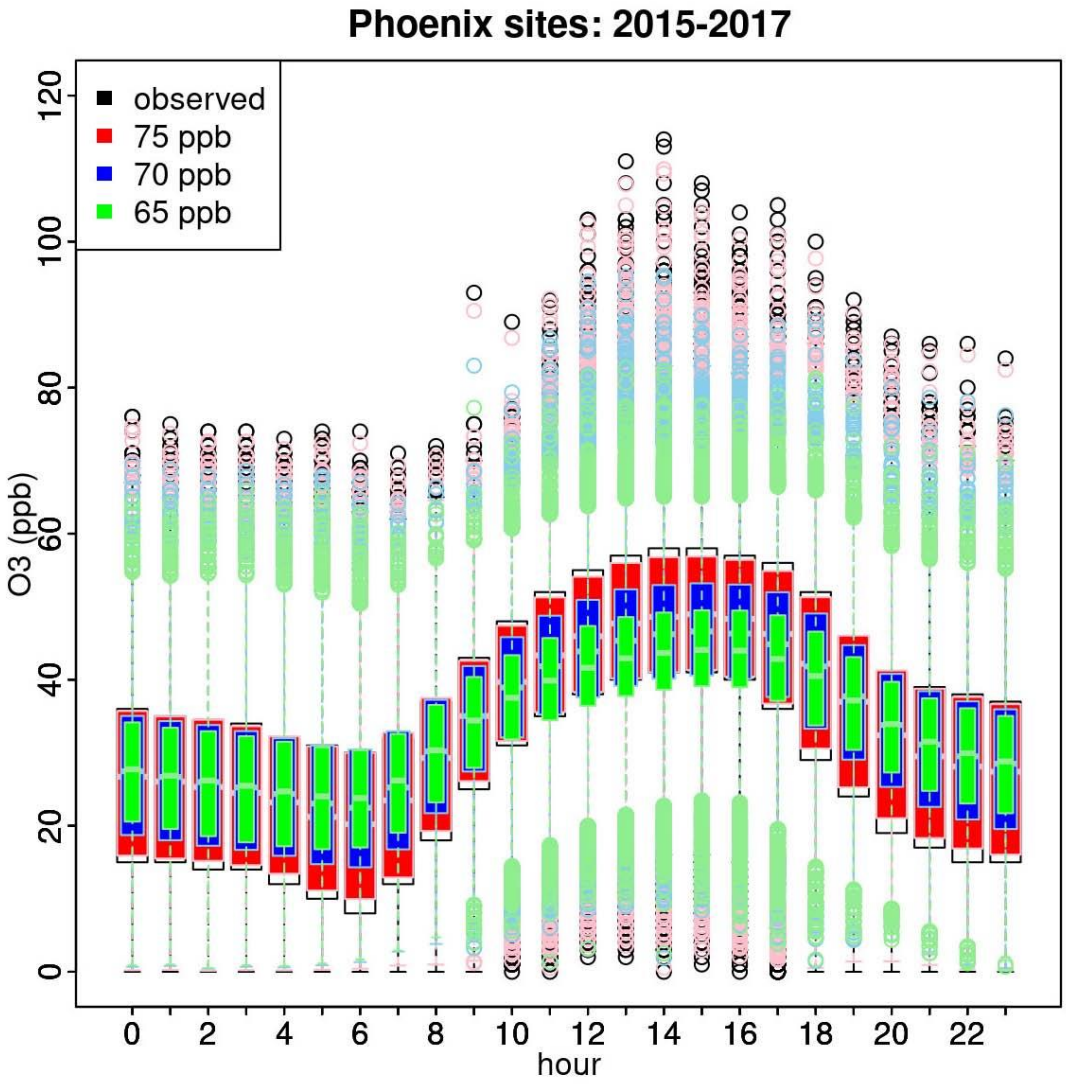


Figure 3C-72. Diurnal distribution of hourly O_3 concentrations at monitoring sites in Phoenix.

Figure 3-8. Figure 3C-72 from US EPA (2019b).

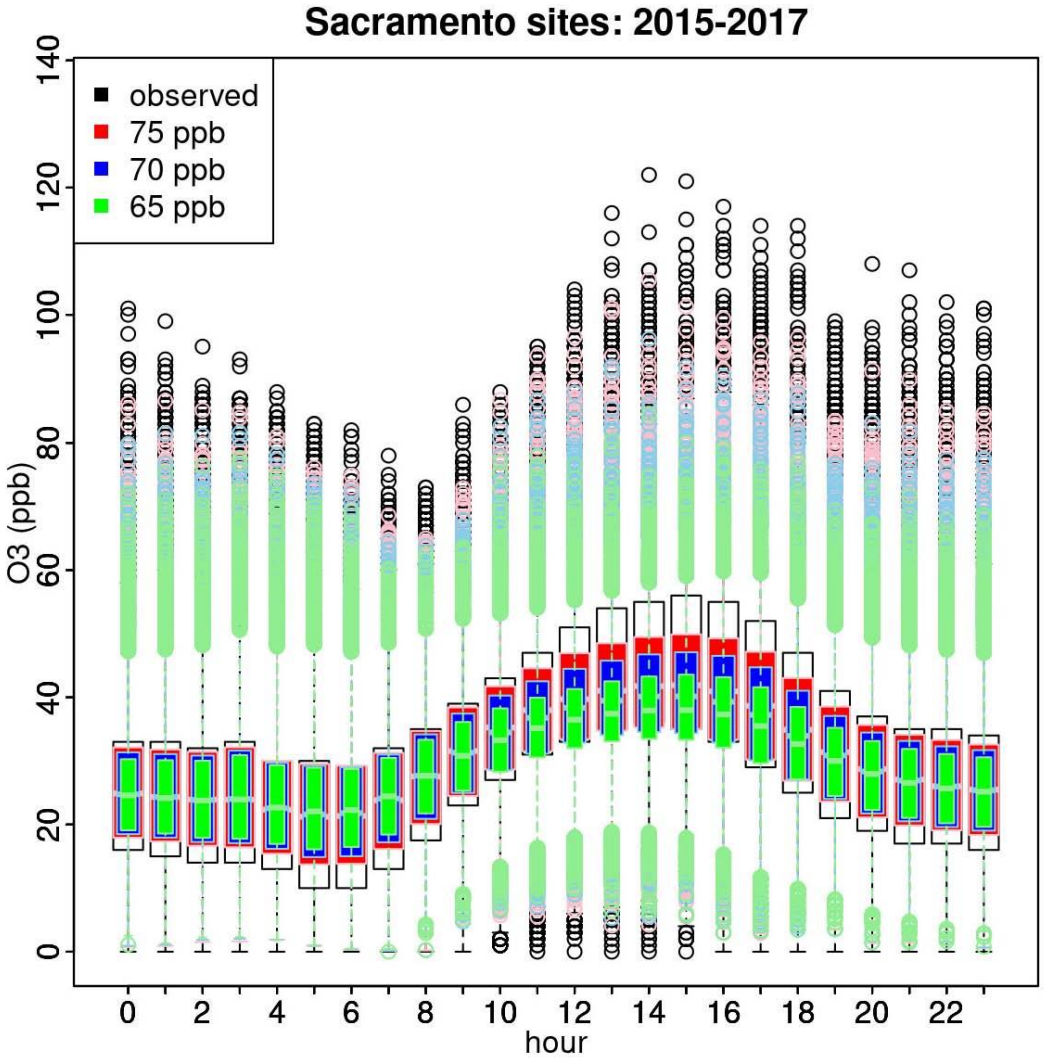


Figure 3C-73. Diurnal distribution of hourly O_3 concentrations at monitoring sites in Sacramento.

Figure 3-9. Figure 3C-73 from US EPA (2019b).

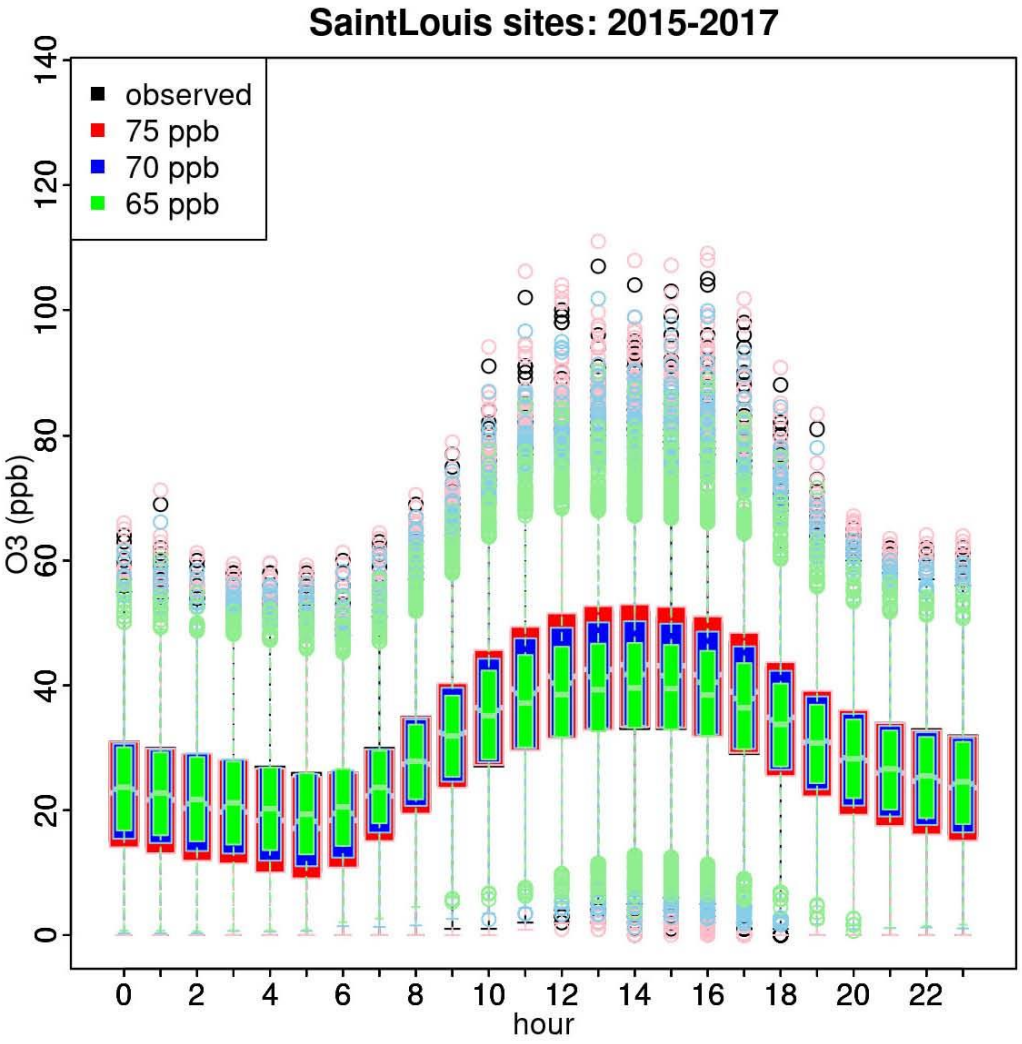


Figure 3C-74. Diurnal distribution of hourly O_3 concentrations at monitoring sites in St. Louis.

Figure 3-10. Figure 3C-74 from US EPA (2019b).

Annual and summer mean and median hourly O_3 concentrations have been used by some researchers to characterize trends, assess human health long-term effects, and evaluate global models. However, because emission reductions result in the high end shifting downward and the low end of the distribution of hourly average O_3 concentrations shifting toward the mid-level values, the average or median values ***increase*** at some sites. There are varying levels of agreement between trends in mean and median concentrations versus different metrics associated with the higher hourly or 8-h average concentrations. Lefohn et al. (2018) reported that trends in the 8-h average metric (an index associated with the high end of the distribution) bear the least

resemblance to trends in the mean and median values with generally less than 50% of sites analyzed having trends in the same direction. Lefohn et al. (2018) reported overall that trends in the mean/median metrics were not representative of the trend behavior of those exposure metrics that focused on the higher end of the distribution. In Fig. 3-11 (reproduced from Lefohn et al., 2018), trend patterns for monthly average concentrations, annual SOMO35, and annual 4th highest daily maximum 8-h concentration (A4MDA8) exposure metrics at a suburban site in Philadelphia, Pennsylvania are compared. The SOMO35 is defined in the EU as the annual sum of the positive differences between the daily maximum 8-h O_3 average value and the cutoff value set at 35 ppb calculated for all days in a year. The monthly average concentrations significantly increased for seven of the 12 months, and were never estimated to decrease, while the SOMO35 and the A4MDA8 metrics, which focused on the higher hourly average concentrations, significantly decreased.

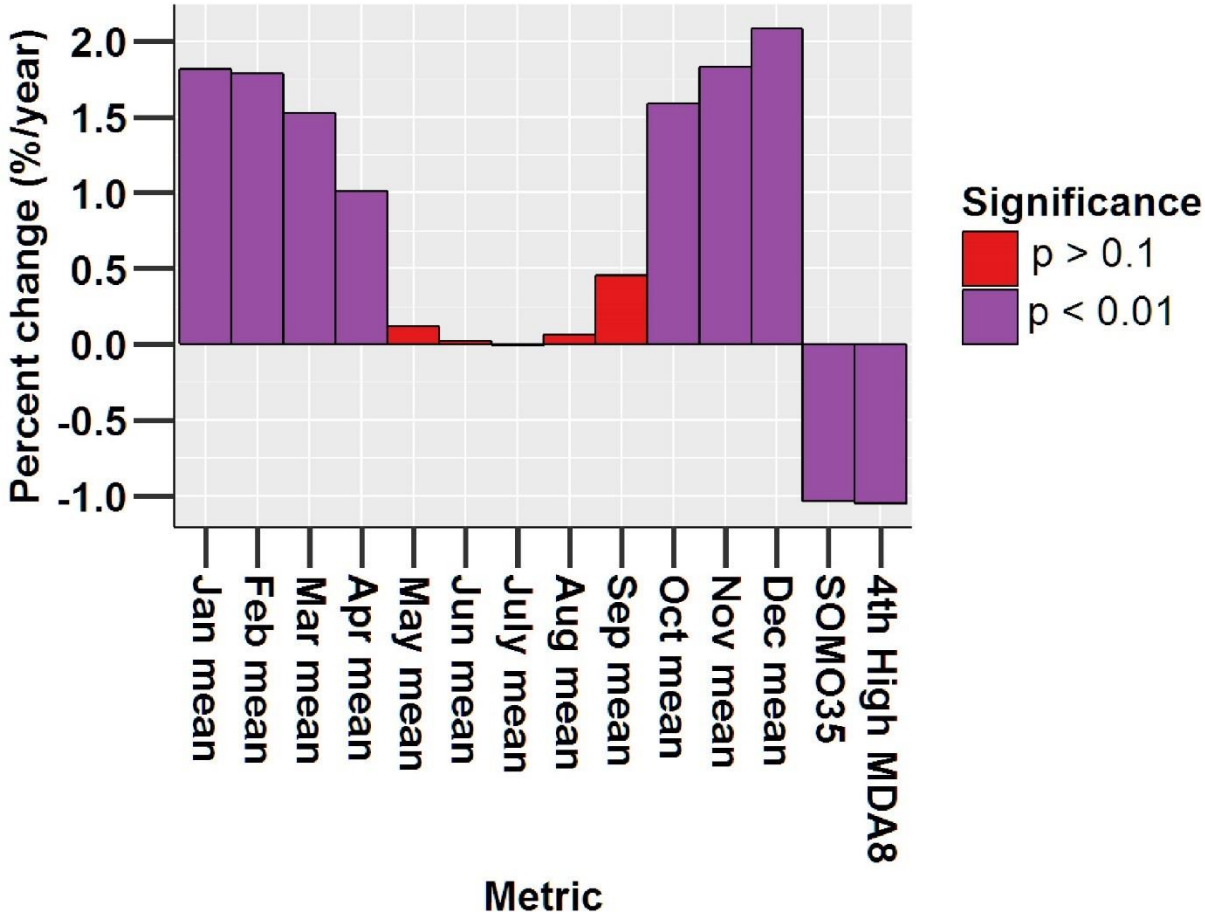


Figure 3-11. The Theil-Sen (%/year) trend in monthly average ozone levels and the annual SOMO35 and 4th highest MDA8 human health metrics (A4MDA8) for a suburban site for 1980-2013 in Philadelphia, Pennsylvania (US EPA AQS ID: 421010024-1). The $p < 0.05$ value was used to determine significance using the Mann-Kendall test. (Source: Lefohn et al., 2018).

For the period 2000 – 2018, Figs. 3-12 and 3-13 illustrate a comparison of the annual 4th highest 8-h daily average concentrations with the annual averages of the hourly average concentrations for 5 sites (Simi Valley in southern CA; Queens, NY; Denali National Park, AK; Voyageurs National Park, MN; and Yellowstone National Park, WY. Note that the Simi Valley site in southern California does not experience exposures that are as high as the design values associated with sites located in San Bernardino County, where in most years the highest design values are experienced in the US. For the period 2016-2018, the design value was 0.111 ppm for two O_3 sites in San Bernardino County. In Fig. 3-12, the two urban sites (i.e., Simi Valley and Queens, NY) experience the highest annual 4th highest daily maximum 8-h average O_3 concentrations in comparison to the three rural National Park sites.

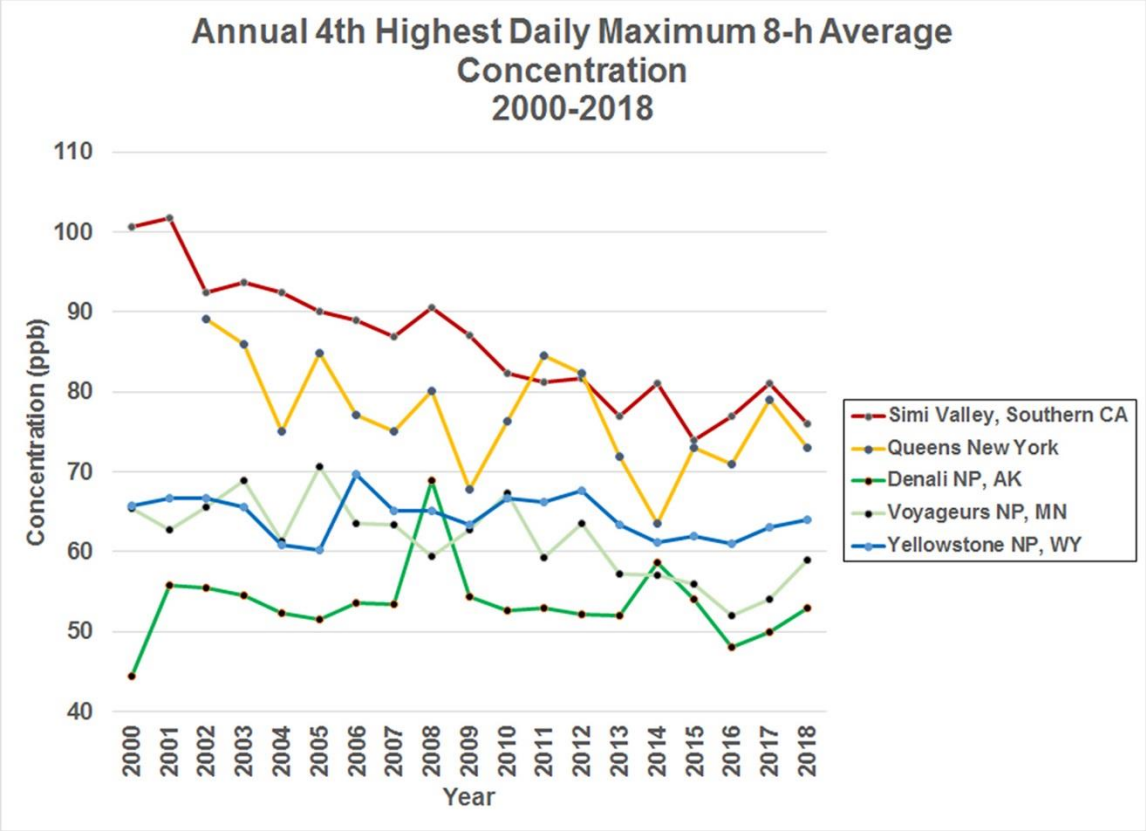


Figure 3-12. The annual 4th highest daily maximum 8-h average O_3 concentration for the period 2000-2018 for Simi Valley, CA (061112002), Queens New York, NY (360810124, Denali National Park, AK (020680003), Voyageurs National Park, MN (271370034), and Yellowstone National Park, WY (560391011).

When the annual average of the hourly average concentrations is calculated for the 5 sites, the Yellowstone National Park site exhibits the highest average concentration values (Fig. 3-13). The annual average values for Voyageurs National Park, Denali National Park, and Simi Valley are similar in value. The Queens, NY site experiences the lowest annual average values, which are increasing over time.

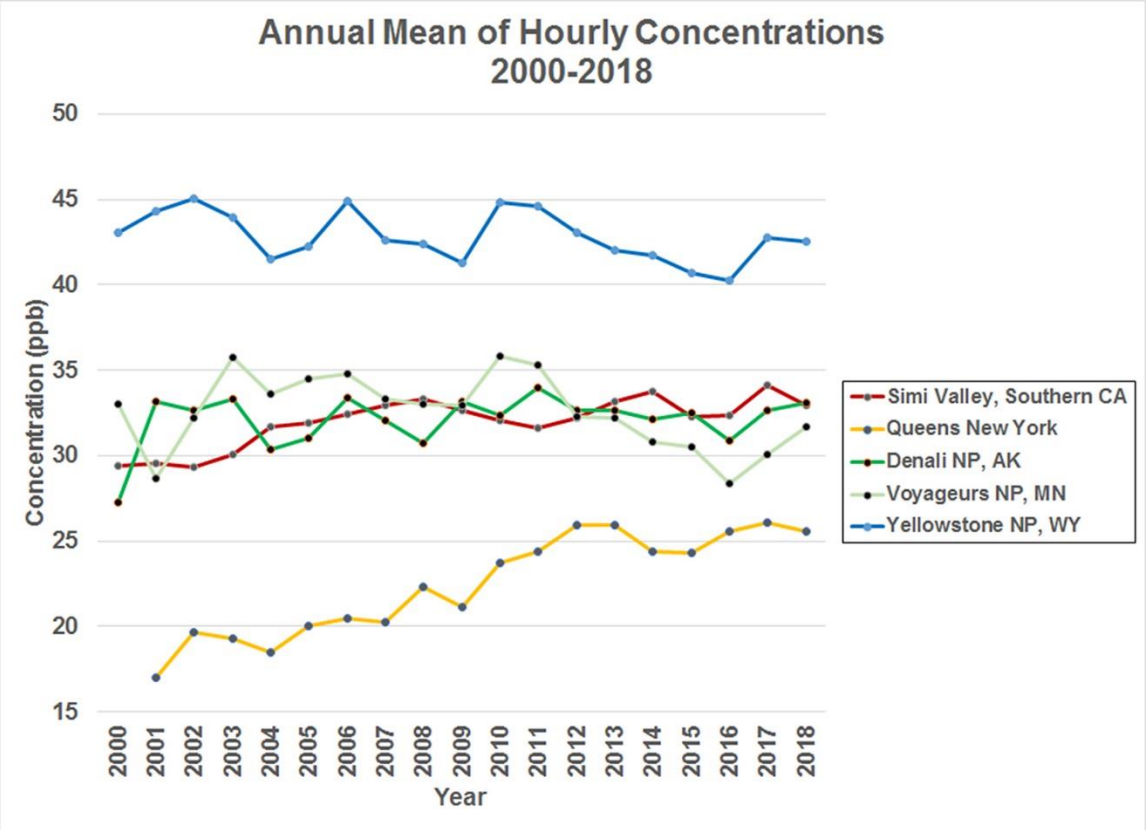


Figure 3-13. The annual mean of the hourly average O_3 concentrations for the period 2000-2018 for Simi Valley, CA (061112002), Queens New York, NY (360810124, Denali National Park, AK (020680003), Voyageurs National Park, MN (271370034), and Yellowstone National Park, WY (560391011).

By calculating a long-term average concentration exposure metric, which combines all hourly values into one number, the highest hourly average concentrations in many cases no longer influence the resulting number because there are many more low- and mid-level values than the higher concentrations. The result of calculating a long-term average is that the values at many of the O_3 monitoring sites having similar values, even though some sites experience elevated hourly O_3 average concentrations and some do not. For example, in the annual mean figure (Fig. 3-13), the ordering of the sites from the highest to the lowest means would appear to be counter intuitive. While the Simi Valley site in southern California experiences the highest 8-h average O_3 exposures of the 5 sites (Fig. 3-12), the annual average concentration for the site is comparable to values for the National Park sites. The three National Park sites in the annual average figures do not experience high 8-h average concentration values comparable to many of the urban sites in the US. Based on the annual mean of hourly average concentrations, the high-elevation Yellowstone National Park site experiences much higher annual average values than any of the other 4 sites. The moderate (i.e., compared to many urban, suburban sites) hourly average O_3 concentrations experienced at Yellowstone National Park (WY) are influenced by frequent occurrences of stratospheric tropospheric transport to the surface, which is a naturally occurring process that contributes to background O_3 levels (Lefohn et al., 2001, 2011, 2012, 2014). Fig 3-14 illustrates for 2007 (modeled background O_3 data provided by EPA) the

relationship between background O_3 levels (defined as apportionment-based USB and referred to as USB_{AB}), stratospheric-tropospheric transport to the surface at the site (STT-S), and the observed ambient hourly average concentrations. The term USB_{AB} will be discussed in Section 3.2. The frequency of STT-S trajectories that arrive at the surface at the site are greatest in the spring but occur throughout the year. Fig. 3-15 illustrates for the entire year (January-December 2006) similar results using Emission Influenced Background (EIB) estimates (see Lefohn et al., 2014) compared with the observed and daily STT-S trajectories. The trajectory model introduced by Wernli and Davies (1997) was used to identify days of high probability for STT trajectories to enhance surface O_3 at specific monitoring sites. The concept of EIB will be discussed as a measure for background O_3 in Section 3.2. An enhanced event occurred on 2 May 2006, when a maximum hourly average O_3 concentration of 89 ppb was measured at 19 UTC (Lefohn et al., 2011). The enhanced event can be seen in Fig. 3-15. There were over 140 STT trajectories that were estimated on that day to reach the surface at the O_3 monitoring site. Škerlak et al. (2019) have described the processes associated with this May 2006 event.

For the period 2000-2014, using data from the TOAR database (Schultz et al., 2017) and the Mann-Kendall nonparametric test, no statistically significant trends at the $p < 0.05$ value were observed at the Yellowstone National Park using either the seasonal 4th highest 8-h daily maximum concentration or the seasonal mean value metrics. Jaffe et al. (2018) in Figure S1 (in their supplement), found no trend at Yellowstone NP for the April-September period for the 4th highest 8-h daily maximum concentration for the 2000-2014 period. It is important that metrics used for assessing trends at sites influenced by natural stratospheric process include the entire 24-h period. For example, when calculating a metric, such as the 4th highest 8-h daily maximum concentration, the entire 24-h period is required to capture the influence of stratospheric events that enhance O_3 concentrations, which at times occur in the late evening or very early morning hours. Reviewing the STT-S daily events for Yellowstone National Park, background O_3 (i.e., USB_{AB}) appears to play a predominant role in influencing the observed ambient levels of O_3.

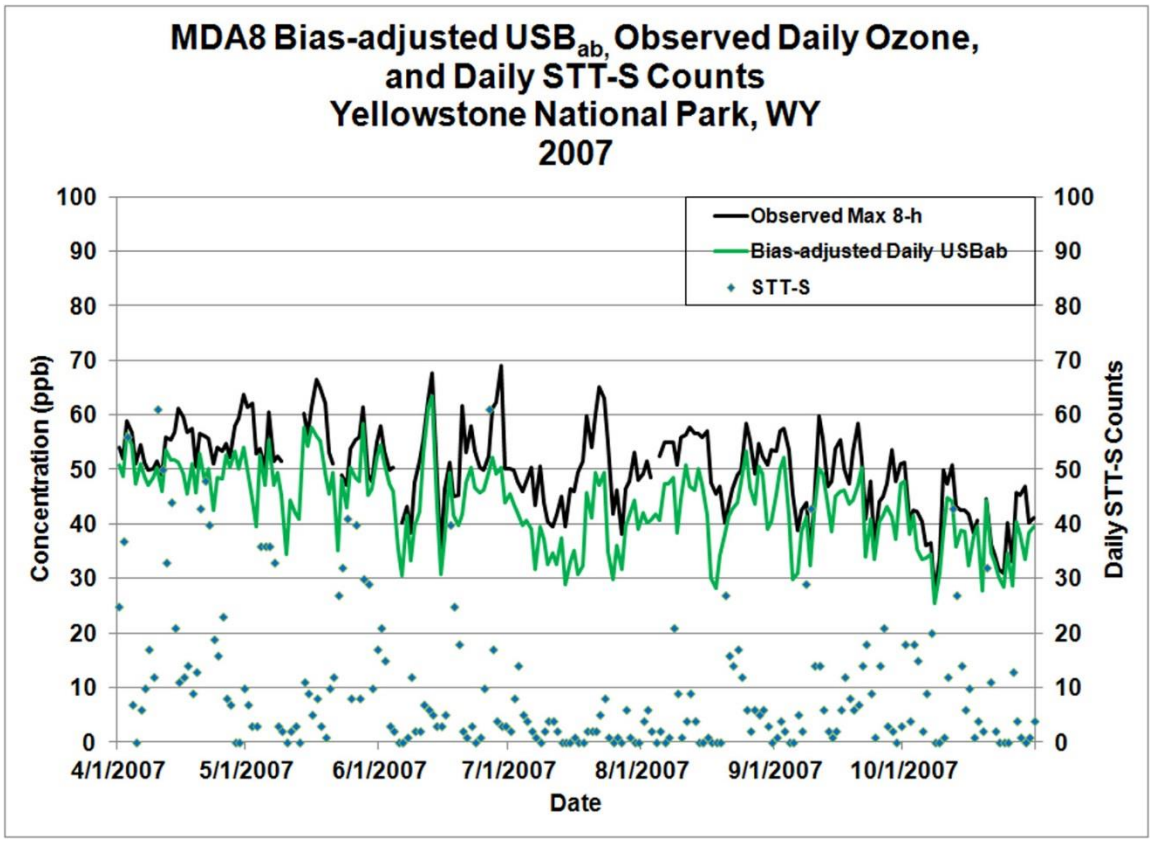


Figure 3-14. A comparison of the observed 8-h daily maximum concentration with the estimated bias adjusted US Background (USB_{AB}) 8-h daily maximum concentrations for Yellowstone National Park (AQS ID 560391011) for April-October 2007. The daily stratospheric-tropospheric transport to surface (STT-S) trajectories, as estimated by Professor Heini Wernli, Institute for Atmospheric and Climate Science, ETH Zurich, Switzerland, are overlaid with the daily O_3 values. Daily USB_{AB} 2007 values provided by the EPA. See Lefohn et al. (2011, 2012, 2014) for details how the STT-S values are estimated.

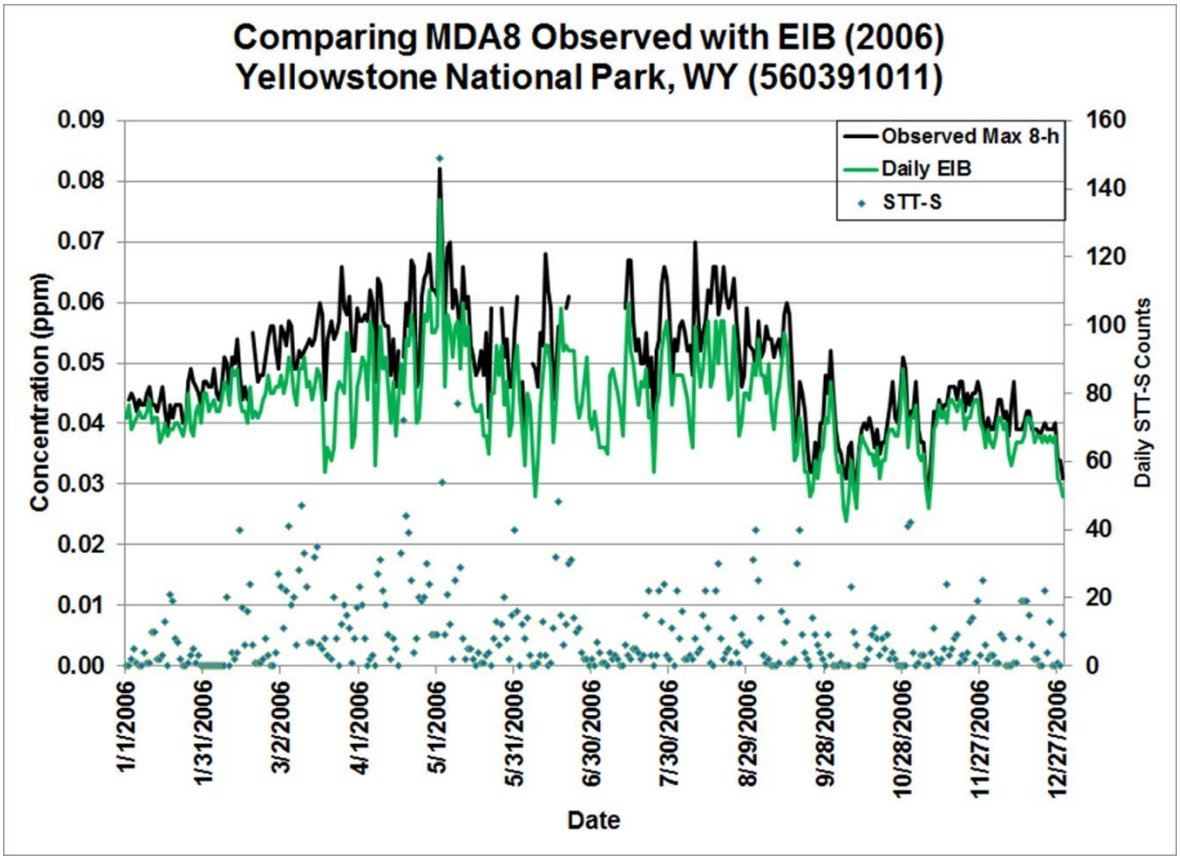


Figure 3-15. A comparison of the observed 8-h daily maximum concentration with the estimated Emissions Influenced Background (EIB) 8-h daily maximum concentrations for Yellowstone National Park (AQS ID 560391011) for January-December 2006. The daily stratospheric-tropospheric transport to surface (STT-S) trajectories, as estimated by Professor Heini Wernli, Institute for Atmospheric and Climate Science, ETH Zurich, Switzerland, are overlaid with the daily O_3 values. See Lefohn et al. (2014) for details how the estimated Emissions Influenced Background (EIB) and STT-S values are estimated. Source: Lefohn et al. (2014).

Using AQS data, information from the TOAR database (see Schultz et al., 2017) indicates that of 406 US sites that reported both annual and 4th highest MDA 8 values, 29% of the sites experienced increasing annual average concentrations for the period 2000-2014, while 65% had no trend, and 6% experienced decreasing annual average concentrations. The nonparametric Mann-Kendall test was used for testing for trends. Thus, a substantial number of the AQS sites had increasing trends using the annual average concentration metric.

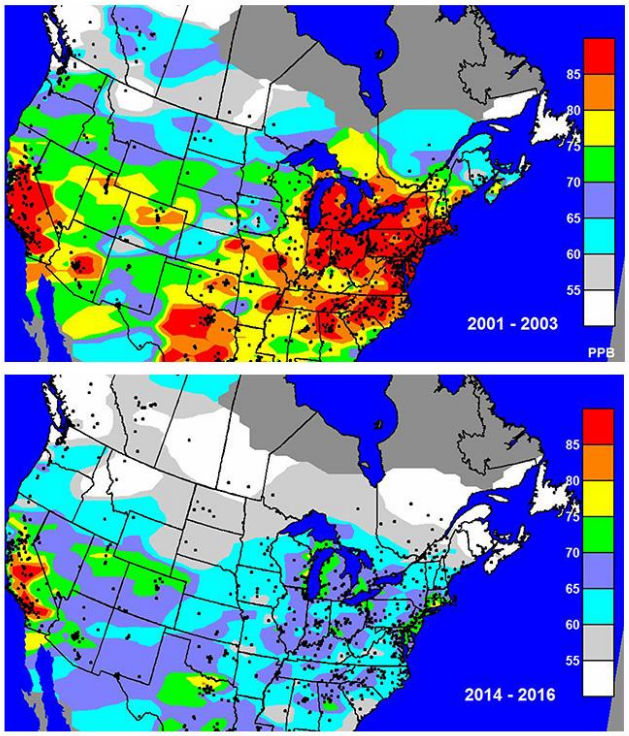
The calculation of the annual mean concentration includes the low, middle, and high hourly average concentrations. Lefohn et al. (1998), Simon et al. (2015) and Lefohn et al. (2017, 2018) have discussed the effects of NO_x emissions on hourly average O_3 concentrations. As pointed out in the draft PA (EPA, 2019b), the greater the amount of NO_x emissions, the greater the frequency of high and low hourly average concentrations for many low-elevation monitoring

sites. Nitrogen oxides scavenge O_3, with the result that the frequency of low hourly average values increase, and the frequency of the higher hourly levels increase due to precursors production of O_3. Simon et al. (2015) discussed the effects of reducing O_3 precursors in the United States on O_3 concentrations. Using daily 8-h average concentrations, the authors reported that decreasing O_3 trends generally occurred in the summer, in less urbanized areas, and at the upper end of the O_3 distribution (i.e., the higher 8-h concentrations). Conversely, ***increasing*** O_3 trends generally occurred in the winter, in more urbanized areas, and ***at the lower end of the O_3 distribution***. The authors noted that increasing fifth percentile trends (i.e., the trends in the lower end of the distribution) were more common in the more highly urbanized areas. Simon et al. (2015) indicate that as anthropogenic NOx emissions have decreased, the O_3 distribution has been compressed (i.e., less frequent high and low values), leading to less spatial and temporal variability. Lefohn et al. (2017, 2018) noted that there is both modeling and observational evidence that the reductions in the frequency of low levels (i.e., shifts of the lower levels upward) are associated with emissions reductions resulting in less O_3 titration by NO_x. Based on this, one would anticipate that as emissions are reduced, that annual O_3 averages would increase, while the highest 8-h average concentrations would decrease. This is exactly what has been reported in the literature (Lefohn et al., 2017, 2018).

As indicated earlier, during the 2015 O_3 rulemaking, EPA believed that both acute and chronic effects could be reduced by reducing the higher hourly average concentrations. As emissions were reduced, the higher part of the distribution of hourly average concentrations moved downwards toward the middle hourly average concentrations. However, if one believes that to eliminate "chronic" (i.e., long-term) effects from O_3 exposures one must decrease annual average concentrations, based on models and empirical data, this cannot be accomplished. By focusing on an emission reduction strategy to decrease the higher hourly average O_3 concentrations, the potential for chronic and acute health and vegetation effects are reduced (Federal Register, 2015).

3.1.2 Patterns of the Changes in When the Highest O_3 Concentrations Occur As Emissions are Reduced

In the US, we have experienced significant reductions in O_3 levels. Figure 3-16 below compares the 3-year average of the annual 4th highest 8-h value between 2001-2003 with 2014-2016. As a result of emission reductions to attain reductions in O_3 exposures, important changes have occurred in the months when the highest O_3 concentrations are observed. Using models, Figure 3C-75 (page 3C-108) through Figure 3C-82 (page 3C-115) display the same information as Figure 3C-67 through Figure 3C-74 in the draft PA but for monthly rather than diurnal distributions. The figures below illustrate the modeling results presented in the draft PA document (EPA, 2019b) for the shifting of the higher mid-range concentrations from the summer season toward the earlier months as emissions are reduced to attain the three scenarios discussed earlier.



Comparing the 3-year average of the annual 4th highest 8-h value for 2001-2003 versus 2014-2016. Analyses provided to AS Lefohn by Tom Dann, Canada.

A.S.L. & Associates, Helena, MT

Figure 3-16. A comparison of the 3-year average of the annual 4th highest 8-h value for the period 2001-2003 with 2014-2016.

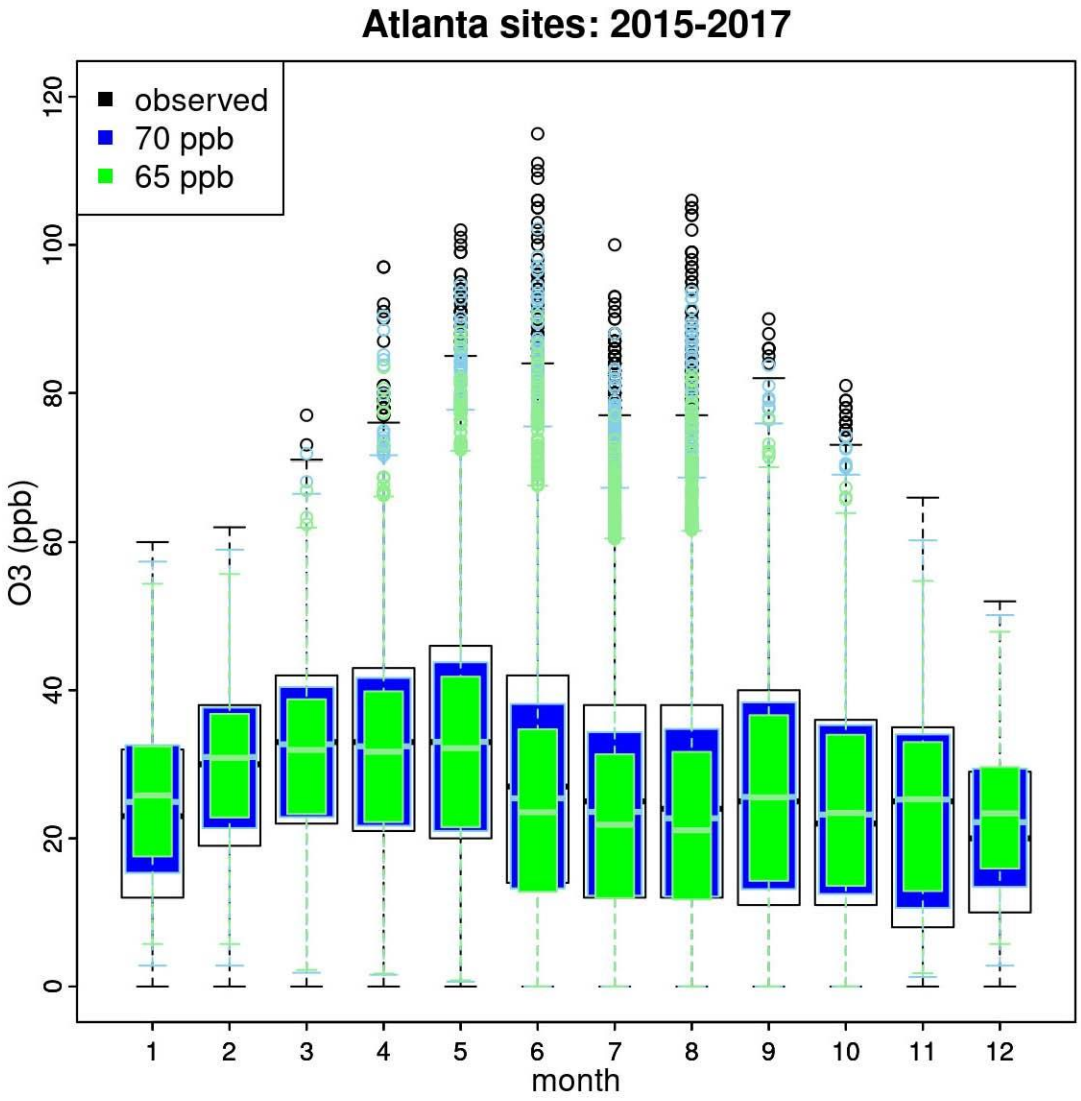


Figure 3C-75. Monthly distribution of hourly O_3 concentrations at monitoring sites in Atlanta.

Figure 3-17. Figure 3C-75 from US EPA (2019b).

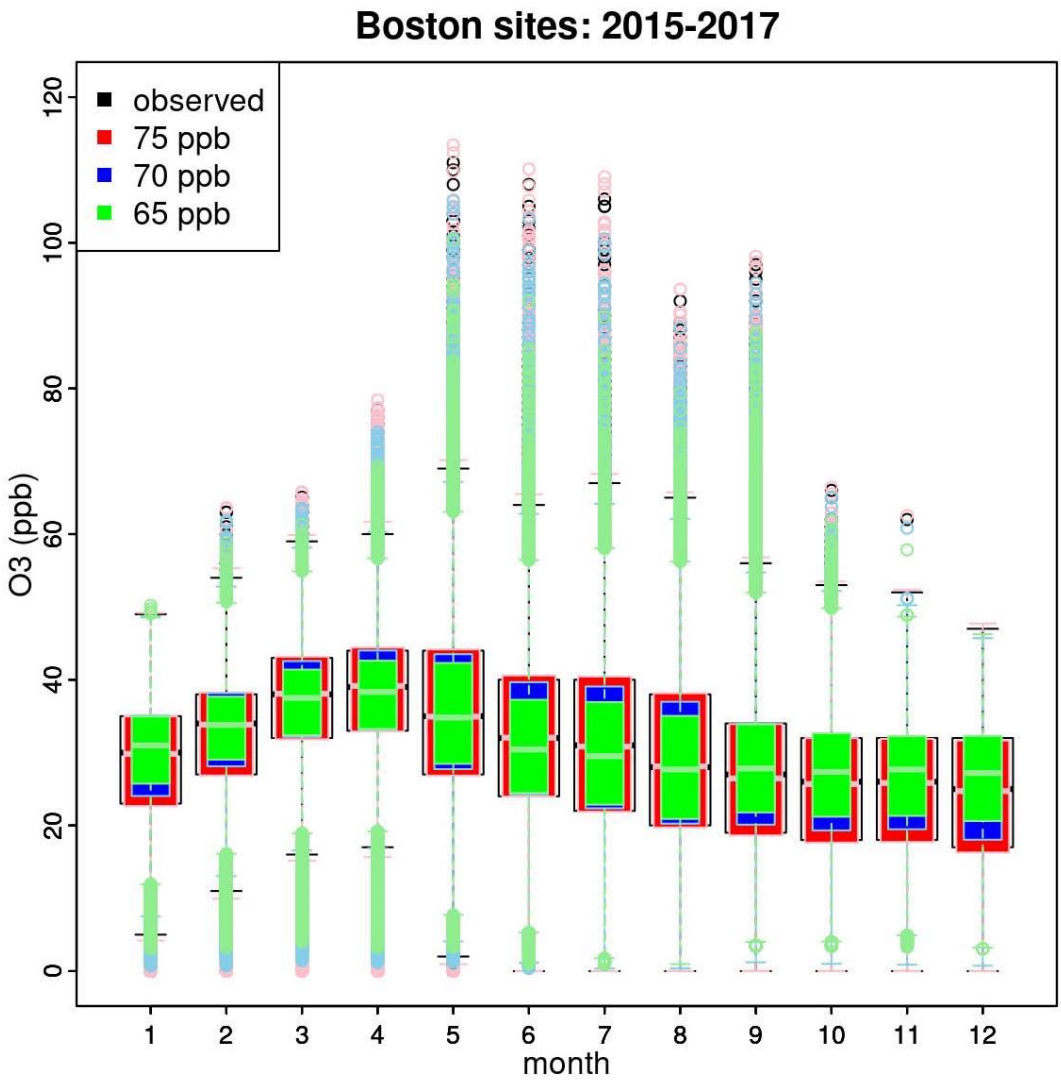


Figure 3C-76. Monthly distribution of hourly O_3 concentrations at monitoring sites in Boston.

Figure 3-18. Figure 3C-76 from US EPA (2019b).

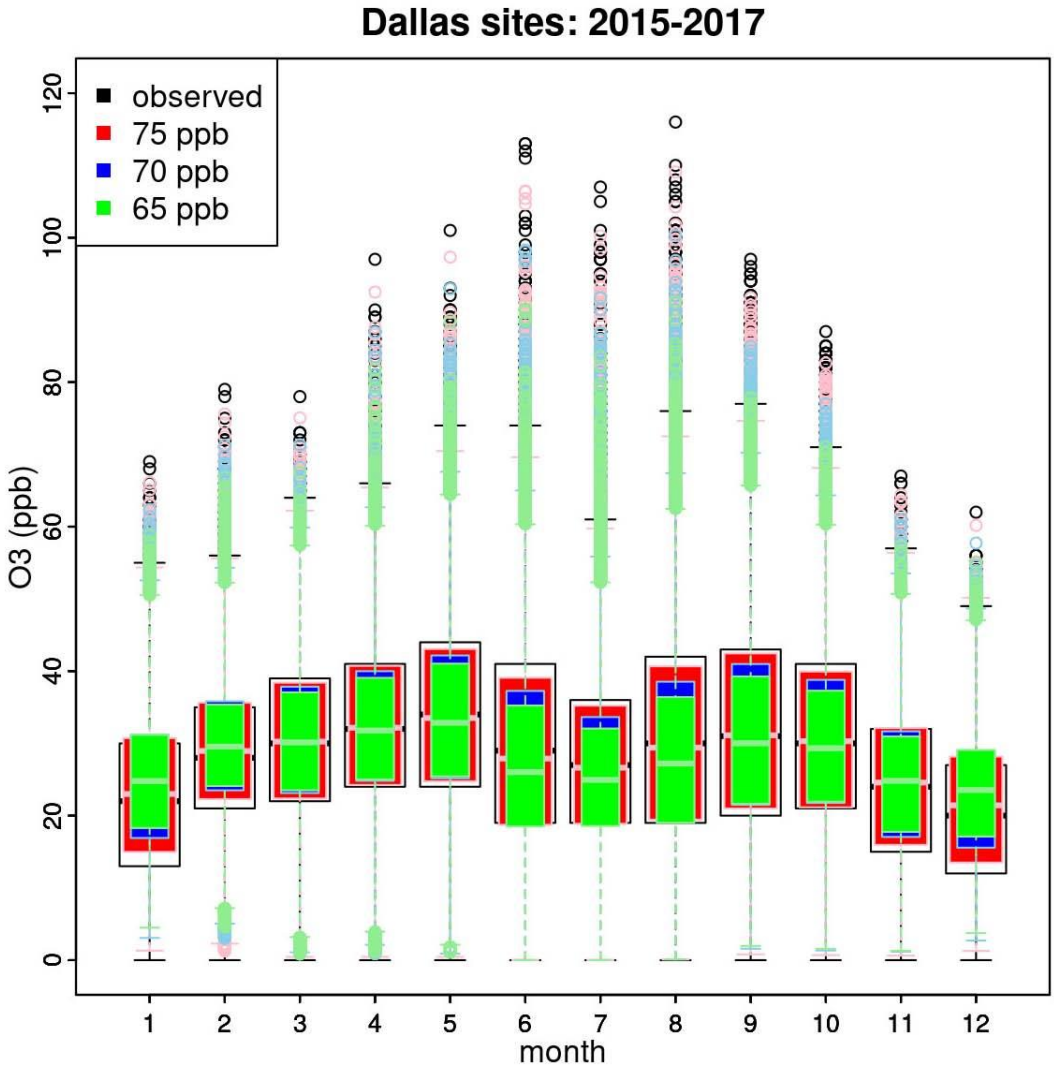


Figure 3C-77. Monthly distribution of hourly O_3 concentrations at monitoring sites in Dallas.

Figure 3-19. Figure 3C-77 from US EPA (2019b).

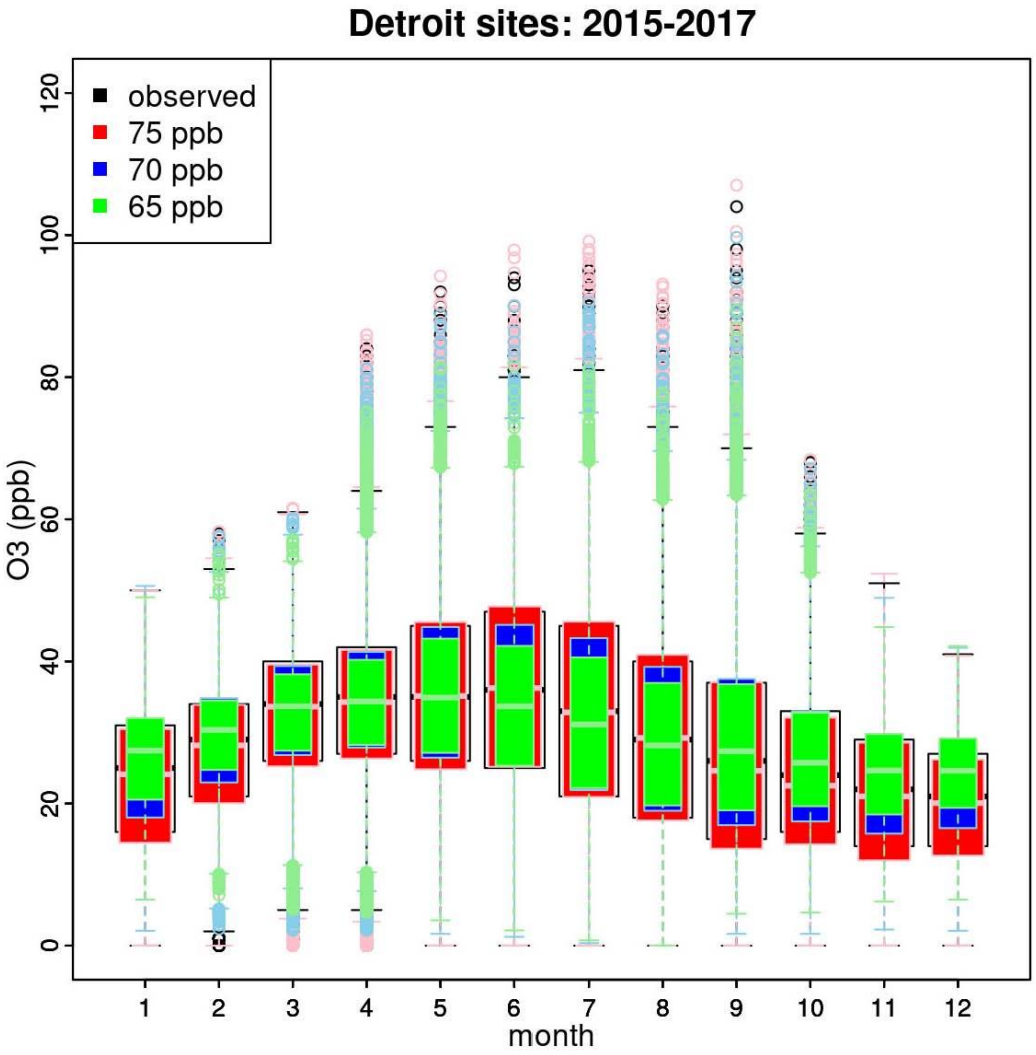


Figure 3C-78. Monthly distribution of hourly O_3 concentrations at monitoring sites in Detroit.

Figure 3-20. Figure 3C-78 from US EPA (2019b).

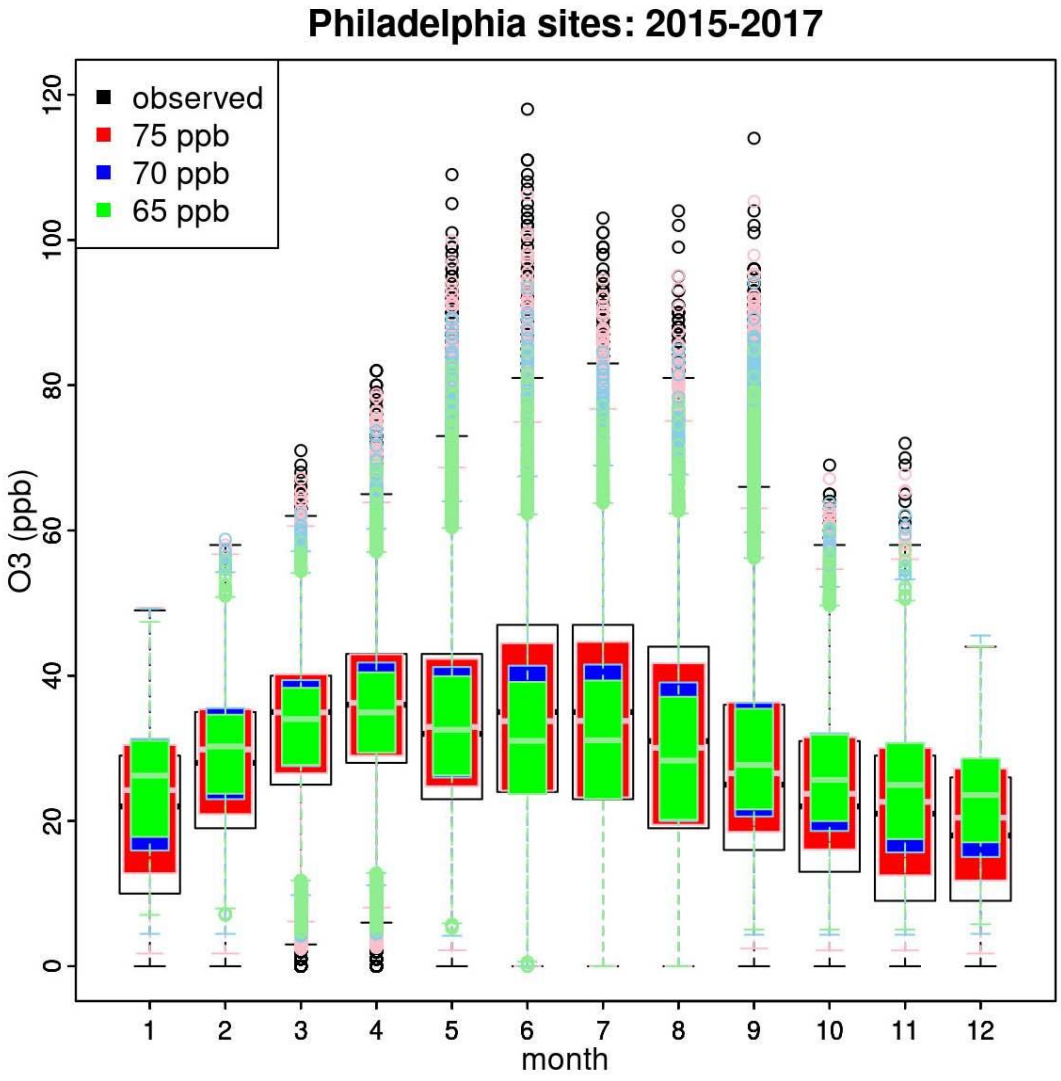


Figure 3C-79. Monthly distribution of hourly O_3 concentrations at monitoring sites in Philadelphia.

Figure 3-21. Figure 3C-79 from US EPA (2019b).

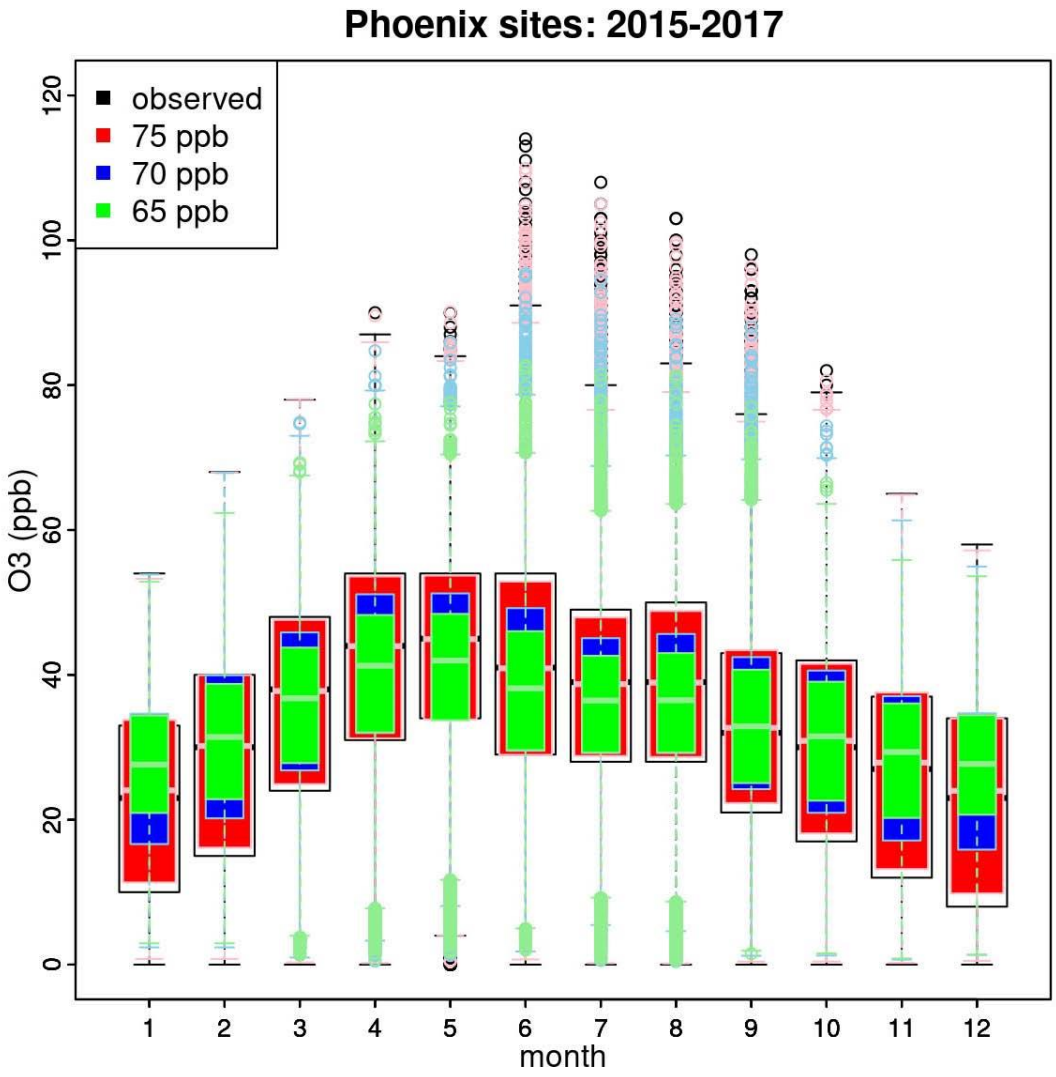


Figure 3C-80. Monthly distribution of hourly O_3 concentrations at monitoring sites in Phoenix.

Figure 3-22. Figure 3C-80 from US EPA (2019b).

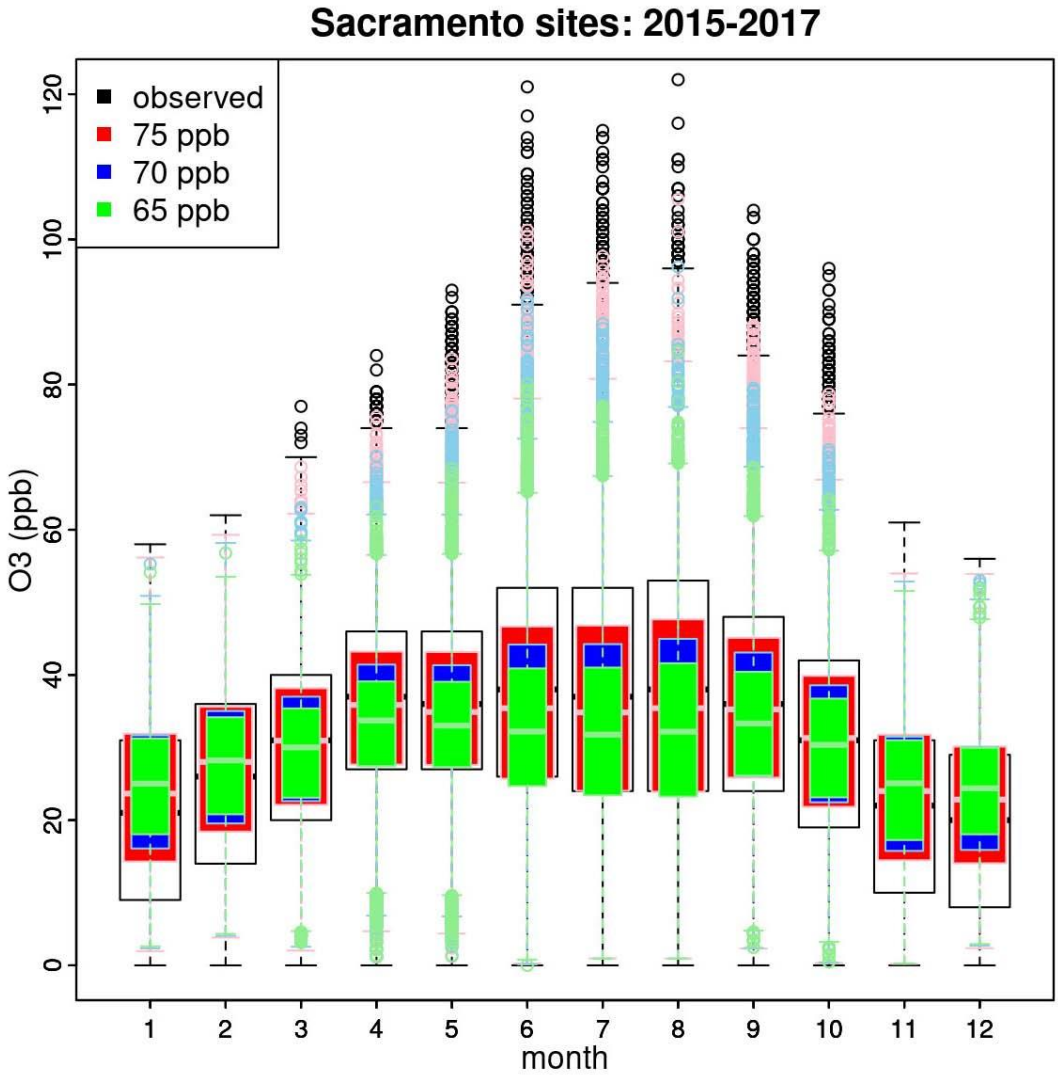


Figure 3C-81. Monthly distribution of hourly O_3 concentrations at monitoring sites in Sacramento.

Figure 3-23. Figure 3C-81 from US EPA (2019b).

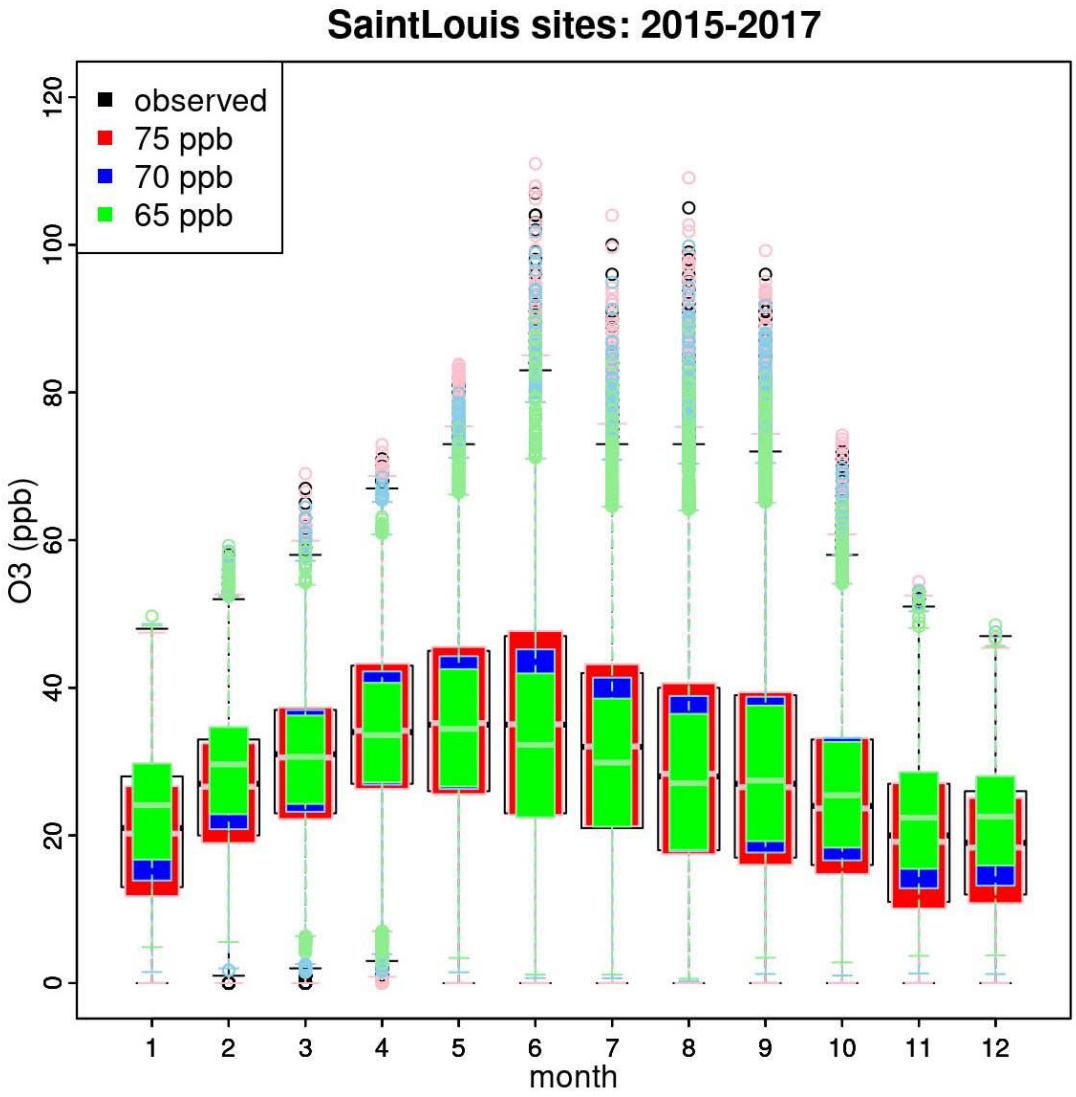


Figure 3C-82. Monthly distribution of hourly O_3 concentrations at monitoring sites in St. Louis.

Figure 3-24. Figure 3C-82 from US EPA (2019b).

Using its models, for the monthly plots for the 8 cities, the EPA notes in the draft PA (EPA, 2019b) on page 3C-99 the following:

> Similar to the diurnal plots, the seasonal distributions become flatter when adjusted to meet the 70 ppb and 65 ppb scenarios, especially on the highest O_3 days. This is due to more O_3 decreases during summer months and more O_3 increases in winter months. The O_3 increases in the winter are consistent with the understanding that solar insolation rates are lower in the winter reducing total photochemical activity and shifting the net

effect of NO_X emissions on O_3 which can both create O_3 through photochemical pathways and destroy O_3 through titration. In addition, the decreases on the highest O_3 days and increases on the lowest O_3 days show a visible compression of the O_3 distribution in these plots, similar to what was seen in the diurnal plots.

The modeling results showed changes for midrange O_3 days for a pattern of shifting higher mid-range O_3 from the summer months to earlier in the year. While in most cities, the highest interquartile O_3 concentrations in the recent conditions occur in the summer months, in many areas the highest interquartile O_3 concentrations shift to spring months (April-May) for the adjustment scenarios. This pattern can be seen in Detroit, Philadelphia, Phoenix, and St. Louis. In the previous analysis in the EPA Risk Exposure Assessment (EPA, 2014b), a similar pattern was observed in Atlanta, Baltimore, Boston, Denver, Los Angeles, New York, Philadelphia, Sacramento, and Washington D.C. This pattern is consistent with a greater contribution from non-U.S. anthropogenic sources at lower projected standard levels than under recent observed conditions. Two of these non-U.S. anthropogenic sources, stratospheric intrusions and international transport, have been shown to peak during the spring months as discussed in the draft Integrated Science Assessment (EPA, 2019a page IS-13).

While the modeling results show the pattern of the shifting of the higher mid-range concentrations from the summer months toward the spring months as emissions are reduced, it is important to explore if modeling predictions are confirmed using actual hourly concentration data from O_3 monitoring sites. Actual data do show that the highest O_3 exposures occur at sites across the US during the springtime (March to mid-June). Using hourly average O_3 data from 57 National Parks, the EPA in the 2014 PA (EPA, 2014c) (Welfare Appendix, page 7A-12) provided the highest 3-month W126 values and the timeframe corresponding to those W126 exposures for the National Parks with O_3 monitors for the period 2006-2010. Table 7A-2 is provided in the pages below. Note that several of the O_3 monitors in the National Parks experienced their highest W126 exposures during the spring months (defined as March, April, May or April, May, June) period. While the months of April, May, and June are not entirely a spring period (half of June is still spring), the time of year when the frequency of stratospheric intrusions (i.e., a natural process) to the surface is greatest at many sites during the March – June window. Lefohn et al. (2011, 2012) reported that stratosphere-to-troposphere transport to the surface (STT-S) frequently coincides with "enhanced" surface O_3 concentrations ($\geq$ 50 ppb) at both high- and low-elevation monitoring sites across the US during specific months, especially the spring. Dr. Heini Wernli, Institute for Atmospheric and Climate Science, ETH Zurich in Switzerland, as a part of our international research team, applied a Lagrangian method, based on the approach introduced by Wernli and Bourqui (2002), to identify stratosphere-to-troposphere transport (STT) events down to the surface (i.e., STT-S events). The trajectory model introduced by Wernli and Davies (1997) was used to identify days of high probability for STT trajectories to enhance surface O_3 at specific monitoring sites. It is important to note that the analysis of stratospheric intrusions and calculation of the SI parameter as described in Lefohn et al. (2011) captures the frequency and vertical penetration of the intrusions; it does not provide information about the O_3 concentration within the intrusion. The O_3 concentration in stratospheric intrusions down to the lower troposphere was expected to be highly variable due to concentration differences in the stratospheric origin and in chemical and mixing processes during the descent.

As noted in Lefohn et al. (2011), this variability can strongly affect any statistical associations between the enhanced hourly average concentrations ≥ 50 ppb used in their analysis and the number of stratospheric intrusions. Thus, Lefohn et al. (2011) chose the coincidence table approach that summarized the frequency of daily intrusions and the daily maximum hourly average O_3 concentrations and applied appropriate statistical tests. For the high-elevation sites in the western and eastern US, the STT-S coincidences occurred most frequently during spring. However, Lefohn et al. (2012) noted that coincidences between STT-S and enhanced O_3 concentrations occurred at times during the summer, fall, and late winter.

Great Smoky Mountains National Park (GRSM) is the most visited National Park in the United States. It is a relatively small park (~210,433 ha), but topographically complex, with an elevational range of 1757 m. The Park is located in parts of North Carolina and Tennessee. The name "Smoky" comes from the natural fog that often hangs over the range and presents as large smoke plumes from a distance. This fog is caused by the vegetation emitting volatile organic carbon chemicals that have a high vapor pressure and easily form vapors at normal temperature and pressure (Naranjo, 2011). The Park has historically been subject to elevated levels of pollutants, including SO_2, O_3, and NO_x. Neufeld et al. (2019) analyzed O_3 trends from 1989 to 2016 for six monitoring sites in and adjacent to GRSM and ranging in elevation from 564m to 2030m. The W126 exposures increased between the years 1989–~2002 and have substantially decreased afterwards. Similar to the pattern described in the modeling results in the draft PA, as emissions were reduced, at most of the six sites analyzed by Neufeld et al. (2019), the maximum 3-month W126 exposures shifted from mid-summer to spring (April–June). Decreases in W126 exposures were correlated with lowered NO_x emissions from regional TVA power plants.

Besides the National Parks, a review of the data in EPA's AQS database indicates that there are many O_3 monitoring sites at both high and low elevations across the US that exhibit highest exposures during the spring months. There are sites where maximum concentrations have shifted from summer to spring months, which confirms the predictions of the models. There are also sites where shifts may not have occurred because the maximum concentrations continue in most years to occur during the spring months. At sites influenced by STT-S, there is a tendency for the highest O_3 exposures to occur during the spring months, but some sites may experience STT-S contributions throughout the year (Lefohn et al., 2011, 2012, 2014). Figs. 3-14 and 3-15, shown previously on pages 50 and 51, illustrate that while the highest O_3 exposures at the high-elevation Yellowstone National Park site may occur during the spring months, enhanced O_3 levels exist throughout the entire year.

Table 7A-2 Ozone Exposure in 57 O_3 Monitors Located in Parks*

Monitor site ID	Park Name	W126					3-Month Timeframe for W126				
		2006	2007	2008	2009	2010	2006	2007	2008	2009	2010
230090102	Acadia National Park	10.59	7.89	7.64	7.02	5.24	MJJ	AMJ	MJJ	MAM	MAM
230090103	Acadia National Park	6.37	6.41	4.72	5.21	4.13	MJJ	AMJ	MJJ	MAM	MAM
311651001	Agate Fossil Beds National Monument	--	8.27	12.76	5.85	--	--	JAS	MJJ	JJA	--
460710001	Badlands National Park	--	--	2.23	2.54	3.85	--	--	JAS	AMJ	JJA
460711001	Badlands National Park	16.74	8.01	--	--	--	JJA	JJA	--	--	--
480430101	Big Bend National Park	11.62	10.60	10.55	8.62	8.47	AMJ	MAM	MAM	MAM	MAM
370110002	Blue Ridge Parkway	9.88	11.46	8.81	4.71	8.19	AMJ	AMJ	AMJ	AMJ	AMJ
490370101	Canyonlands National Park	18.06	16.93	17.06	12.23	13.24	MJJ	MJJ	AMJ	MAM	AMJ
250010002	Cape Cod National Seashore	13.47	13.16	12.89	5.25	7.03	MJJ	MJJ	MJJ	AMJ	MJJ
350153001	Carlsbad Caverns National Park	--	8.65	17.50	11.37	7.09	--	AMJ	AMJ	MJJ	AMJ
160310001	City of Rocks National Reserve	--	--	--	--	6.02	--	--	--	--	JJA
80771001	Colorado National Monument	--	11.61	15.04	4.13	8.75	--	JJA	MJJ	JAS	AMJ
450790021	Congaree National Park	12.31	10.78	9.45	3.97	6.32	MAM	MAM	MAM	FMA	MAM
450210002	Cowpens National Battlefield	14.30	7.87	16.05	3.24	8.81	MJJ	AMJ	JJA	FMA	MAM
160230101	Craters of the Moon National Monument	--	10.17	10.88	5.68	7.82	--	JJA	MJJ	MAM	JAS
210131002	Cumberland Gap National Historical Park	--	18.36	10.12	3.58	7.31	--	MJJ	MJJ	MJJ	MJJ
60270101	Death Valley National Park	29.18	32.55	25.57	15.30	10.61	MJJ	MJJ	MJJ	JJA	JAS
560111013	Devil's Tower National Monument	--	--	7.09	5.42	5.44	--	--	JAS	JAS	JJA
490471002	Dinosaur National Monument	--	10.33	13.34	8.39	13.80	--	MJJ	MJJ	MJJ	MJJ
300298001	Glacier National Park	2.90	2.29	3.98	3.53	2.44	JJA	MAM	MAM	AMJ	AMJ
300351001	Glacier National Park	--	--	--	4.91	3.93	--	--	--	MJJ	MJJ
40058001	Grand Canyon National Park	21.66	18.68	17.02	10.10	14.95	MJJ	AMJ	AMJ	JJA	AMJ
320330101	Great Basin National Park	15.54	15.79	16.94	10.19	11.44	JJA	MJJ	MJJ	AMJ	AMJ
370870036	Great Smoky Mountains National Park	11.46	13.35	11.50	4.59	7.89	AMJ	AMJ	AMJ	AMJ	AMJ
470090102	Great Smoky Mountains National Park	12.97	12.69	10.44	5.31	10.27	AMJ	MAM	AMJ	MAM	MAM
471550101	Great Smoky Mountains National Park	18.87	20.66	14.15	9.03	15.16	AMJ	AMJ	MJJ	MAM	MAM
471550102	Great Smoky Mountains National Park	19.59	23.51	16.23	7.32	11.94	MJJ	JJA	MJJ	MJJ	ASO

Source: US EPA (2014c).

Monitor site ID	Park Name	W126					3-Month Timeframe for W126				
		2006	2007	2008	2009	2010	2006	2007	2008	2009	2010
180890022	Indiana Dunes National Lakeshore	8.79	12.21	3.66	2.42	3.91	JJA	AMJ	JAS	MJJ	JJA
60650008	Joshua Tree National Park	24.36	19.97	27.43	19.66	23.39	AMJ	AMJ	MJJ	AMJ	AMJ
60651004	Joshua Tree National Park	--	26.37	30.05	18.81	20.47	--	MJJ	AMJ	JJA	JJA
60719002	Joshua Tree National Park	55.48	52.46	50.99	39.93	43.92	MJJ	MJJ	JJA	JJA	JJA
60893003	Lassen Volcanic National Park	18.97	15.10	18.98	7.64	9.63	JAS	JJA	MJJ	JJA	JAS
80830101	Mesa Verde National Park	23.44	17.57	13.41	15.05	11.94	MJJ	MJJ	AMJ	JJA	AMJ
60711001	Mojave National Preserve	--	28.50	38.92	19.91	19.39	--	MJJ	MJJ	JAS	JJA
530530012	Mount Rainier Wilderness	3.19	3.30	1.18	2.20	1.86	MAM	MAM	JAS	FMA	MAM
530090016	Olympic National Park	--	--	--	--	0.52	--	--	--	--	JAS
530091004	Olympic National Park	--	0.28	0.93	--	--	--	JAS	JAS	--	--
482731001	Padre Island National Seashore	--	8.19	3.66	--	--	--	AMJ	AMJ	--	--
40170119	Petrified Forest National Park	19.16	16.60	19.40	9.04	12.71	AMJ	AMJ	AMJ	AMJ	AMJ
60690003	Pinnacles National Monument	17.14	14.85	19.78	11.41	9.79	JAS	AMJ	MJJ	JAS	JAS
40190021	Saguaro National Park	19.57	17.06	20.13	11.01	15.31	MJJ	MJJ	AMJ	MAM	AMJ
360910004	Saratoga National Historical Park	6.68	10.38	9.26	5.40	5.98	JJA	MJJ	AMJ	MAM	MJJ
311570005	Scotts Bluff National Monument	--	--	--	--	6.20	--	--	--	--	JJA
61070006	Sequoia-Kings Canyon National Park	50.09	53.38	57.24	29.13	26.93	JJA	JJA	JJA	JAS	JAS
61070009	Sequoia-Kings Canyon National Park	66.07	62.88	56.91	55.51	53.79	JAS	JJA	MJJ	JAS	JAS
511130003	Shenandoah National Park	16.43	14.40	12.07	7.63	10.84	AMJ	AMJ	AMJ	MAM	JAS
380070002	Theodore Roosevelt National Park	7.71	5.54	5.55	3.95	4.19	JAS	JJA	AMJ	AMJ	AMJ
380530002	Theodore Roosevelt National Park	9.45	6.29	6.31	4.22	5.17	JJA	JJA	MJJ	AMJ	MAM
40070010	Tonto National Monument	26.39	23.24	25.40	13.67	16.90	MJJ	MJJ	AMJ	AMJ	AMJ
271370034	Voyageurs National Park	5.33	5.19	3.86	4.94	7.66	AMJ	AMJ	MAM	MAM	MAM
460330132	Wind Cave National Park	20.52	12.20	5.92	5.75	5.61	JJA	JJA	JJA	JJA	JAS
560391011	Yellowstone National Park	12.98	9.96	8.84	7.63	11.54	AMJ	AMJ	MAM	MAM	AMJ
60430003	Yosemite National Park	33.78	29.68	42.51	25.70	27.34	JJA	MJJ	JJA	JAS	JAS
60431002	Yosemite National Park	--	12.60	10.03	--	--	--	AMJ	MJJ	--	--
60431003	Yosemite National Park	--	11.61	--	--	--	--	JAS	--	--	--
60431004	Yosemite National Park	--	6.95	15.52	6.58	9.43	--	MJJ	JJA	JAS	JAS
60431005	Yosemite National Park	--	--	27.83	5.18	14.28	--	--	JAS	JAS	JAS

*Nine parks have more than 1 monitor

Source: US EPA (2014c).

3.2 Background Ozone

3.2.1 Why is the Quantification and Spatial Distribution of Background O_3 Important?

It appears from reading both the draft ISA (EPA, 2019a) and the draft PA (EPA, 2019b) that the EPA is focused on answering the following question:

- How much of the current ozone can be attributed to sources other than U.S. anthropogenic sources?

On page 2-23 in the draft PA, the authors note:

> In this review, as in past reviews, the EPA generally characterizes O_3 concentrations that would exist in the absences of U.S. anthropogenic emissions as U.S. background (USB). ***An alternative phrasing is the O_3 concentrations created collectively from global natural sources and from anthropogenic sources existing outside of the U.S as USB. Such a definition helps distinguish the O_3 that can be controlled by precursor emissions reductions within the U.S. from O_3 originating from global natural and foreign precursor sources that cannot be controlled by U.S. regulations (draft ISA, section 1.2.2.*** (emphasis added).

A great deal of the focus by EPA in the modeling effort (page 1-49 of the draft ISA (EPA, 2019a)) appears to be on answering the above question as noted below:

- Applications of chemical transport models (CTMs) to estimate USB ozone have found that USB concentrations are relatively constant with increasing total ozone concentration, indicating that days with higher ozone concentrations generally occur because of higher U.S. anthropogenic contributions (Dolwick et al., 2015).

- Based on these considerations, this section emphasizes USB on days with high ozone concentration as the most relevant for discussing USB ozone, and wherever possible, the focus is on estimates of USB under these conditions because they are most relevant for evaluating the potential for a role of USB ozone in contributing to the highest ozone concentrations. Discussion of seasonal and monthly means of hourly data are also included because longer averaging times are relevant to assessments of health and ecological effects.

Continuing to focus on the contribution of USB on high O_3 days, page 1-57 in the draft ISA (EPA, 2019a) states:

- There is consistent evidence across several studies using different background measurement approaches that USB or other background concentration estimates on most days with high ozone concentrations have been generally predicted to be similar to or smaller than seasonal mean USB ozone estimates in the eastern U.S. and in urban and low-elevation areas of the western U.S., and an inverse

relationship between relative USB contribution and total ozone concentration in these areas has been consistently predicted. This contrasts with high-elevation locations in the western U.S., where USB and NAB have been consistently predicted to increase with total ozone concentration.

Further, in the draft PA (2019b), this focus continues on page 2-60 of the draft PA where the Agency provides reasons for its interest in background O_3 when it notes in its summary bullets:

- It is important to recognize that exceedances of the existing standards have a substantially higher model-predicted USA (anthropogenic) contribution than other days in both the West and the East.

- International contributions, in most places, are lowest during the season with the most frequent O_3 exceedances. Except for the near-border areas, the International contribution requires long-distance transport that is most efficient in Spring.

- The USA contributions that drive exceedances generally peak in summer. In this typical case, USB is overwhelmingly from Natural sources. The most notable exception is near the Mexico border where the modeling indicates that a combination of Natural and Canada/Mexico contributions can lead to total USB between 60-80 ppb on specific days, which is consistent with the previous O_3 Policy Assessment.

While it appears to that the Agency is focused on how much of the current O_3 can be attributed to sources other than U.S. anthropogenic sources on days when ambient levels exceed the standard, there are other considerations in the rulemaking process that are of equal importance to quantifying background O_3. As discussed above, EPA states on page 1-49 of the draft ISA (EPA, 2019a), that background seasonal and monthly means of hourly data are also included because longer averaging times are relevant for assessments of health and ecological effects. However, this statement is inaccurate. In many cases, assessment of human health and ecological effects are not based on longer averaging times. For the vegetation related W126 exposure index, which is not an average exposure metric, hourly average concentrations are weighted and accumulated over a specific period for assessing vegetation effects. Hourly average background O_3 concentrations contribute to the observed concentrations and therefore, contribute to the cumulative risk. For some human health risk assessments, daily maximum 8-h average concentrations are used in a time series. Daily maximum 8-h average concentrations contain background O_3 concentrations, which contribute to the estimated human health risk.

Background O_3 concentrations in the low- and mid-level part of the distribution of concentrations make up a large fraction of the total O_3 levels and the lower and mid-level concentrations influence mortality and morbidity risk estimates. It is important to quantify the importance of background O_3 in the low- and mid-range concentrations. Fig. 3-25 (reproduced from Fig. 9-8 on Page 9-32 from the 2014 HREA (EPA, 2014b)) illustrates the percent reduction in exposures and risks after just meeting alternative standards relative to just meeting the 2008 NAAQS of 75 ppb. In this plot, each row represents one of the key analytical results and each

column gives the results for 2007 and 2009 for each urban study area. The scales are the same between analyses, and as such, it is informative to examine both the overall patterns of change between alternative standards, and the absolute value of the percent reductions in risk metrics between analyses. The top row is the Exposure > 60 ppb; the second row is the Lung Function Risk ($dFEV_1$ > 10%); the third row is Mortality; the fourth row is Hospital Admissions. The risks associated with mortality and hospital admissions are much less than the risks associated with Exposure > 60 ppb and Lung Function Risk ($dFEV_1$ > 10%). This is because mortality and hospital admission risk metrics are based on non-threshold, approximately linear C-R functions, and therefore are sensitive to changes in O_3 along the full range of O_3 concentrations (page 9-30 of the 2014 Health REA (EPA, 2014b)). As explained by the Agency (EPA, 2014b), because O_3 in the lower concentration range may shift upward as the result of NOx emission reductions, this can lead to increases in risk on some days, which can lead to a net increase or decrease in risk over the entire year, depending on whether the days with increased risk exceed days with decreased risk (generally due to a preponderance of days with lower O_3 concentrations). Fig. 3-3-26 illustrates the percent of short-term mortality attributable to O_3 concentrations in the 25-55 ppb range for 2007. The data to create the figure were obtained from EPA (2014d) in Fig. 7-B1 on page 7-B3. In some cases, 90% or more of the accumulated risk is associated with the mid-range concentrations for cities across the U.S. The different colors represent the different standard scenarios considered in the 2014 Health REA (EPA, 2014b). Results shown from the 2014 Heath REA (EPA, 2014b) were similar for all 12 cities used in the 2014 epidemiological risk analyses. The mid-range concentrations (25-55 ppb) as emissions are reduced is where background O_3 will predominate. Fig. 3-27 illustrates the contribution of background to ambient levels of O_3 for 2006 in Houston, Texas (see Lefohn et al., 2014 for further details). A large percentage of the observed concentrations in the 25-55 ppb range are associated with background O_3 at the measured levels in 2006. As indicated in earlier discussions in this document, as emissions are reduced, for some sites, a compression of the distribution of concentrations shifts the lower concentrations upward and the higher concentrations downward. Thus, as shown in Fig. 3-27, background O_3 concentrations will be expected to increase their domination of the cumulative mortality health risk estimates.

Background O_3 concentrations become more and more important in influencing ambient levels as emission reductions are implemented. It is recognized by the Courts that NAAQS O_3 levels are set to protect public health and welfare and that background O_3 is not a consideration in setting these levels. In the draft PA (page 1-11), the EPA notes the following:

> In the August 2019 decision, the court additionally addressed arguments regarding considerations of background O_3 concentrations, and socioeconomic and energy impacts. With regard to the former, the court rejected the argument that the EPA was required to take background O_3 concentrations into account when setting the NAAQS, holding that the text of CAA section 109(b) precluded this interpretation because it would mean that if background O_3 levels in any part of the country exceeded the level of O_3 that is requisite to protect public health, the EPA would be obliged to set the standard at the higher nonprotective level. Thus, the court concluded that EPA did not act unlawfully or arbitrarily or

capriciously in setting the 2015 NAAQS without regard for background ozone.

However, for practical purposes, if the standard were to be set at a level that ambient O_3 levels consisted of almost all background O_3, then the setting of such a level under these circumstances would not be a standard, but a goal that would not necessarily be achievable. By understanding the range of hourly average background O_3 concentrations, scientists can better place into perspective their human health and vegetation results and ask the question: How can my results be valid if the exposures that I have used are at or below background O_3 levels? It is important to note that the range of background O_3 concentrations at a specific site is different than the range at other monitoring sites in the same location. Each site is unique. Thus, quantifying the range of background O_3 concentrations at a specific location may be more relevant than determining the range of O_3 concentrations on a large geographic regional scale.

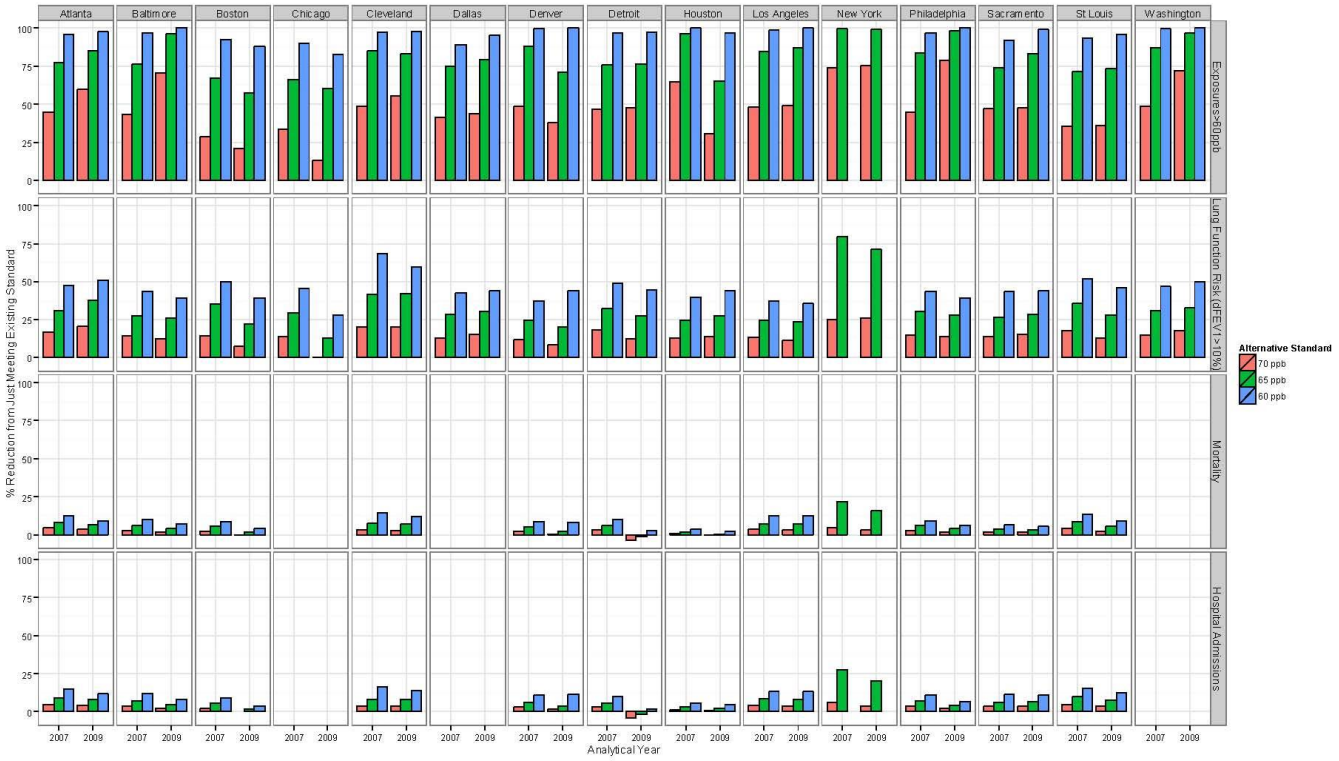


Figure 9-8. Comparison of the Percent Reduction in Key Risk Metrics for Alternative Standard Levels Relative to Just Meeting the Existing 75 ppb Standard.

Figure 3-25. Figure 9-8 from US EPA (2014b).

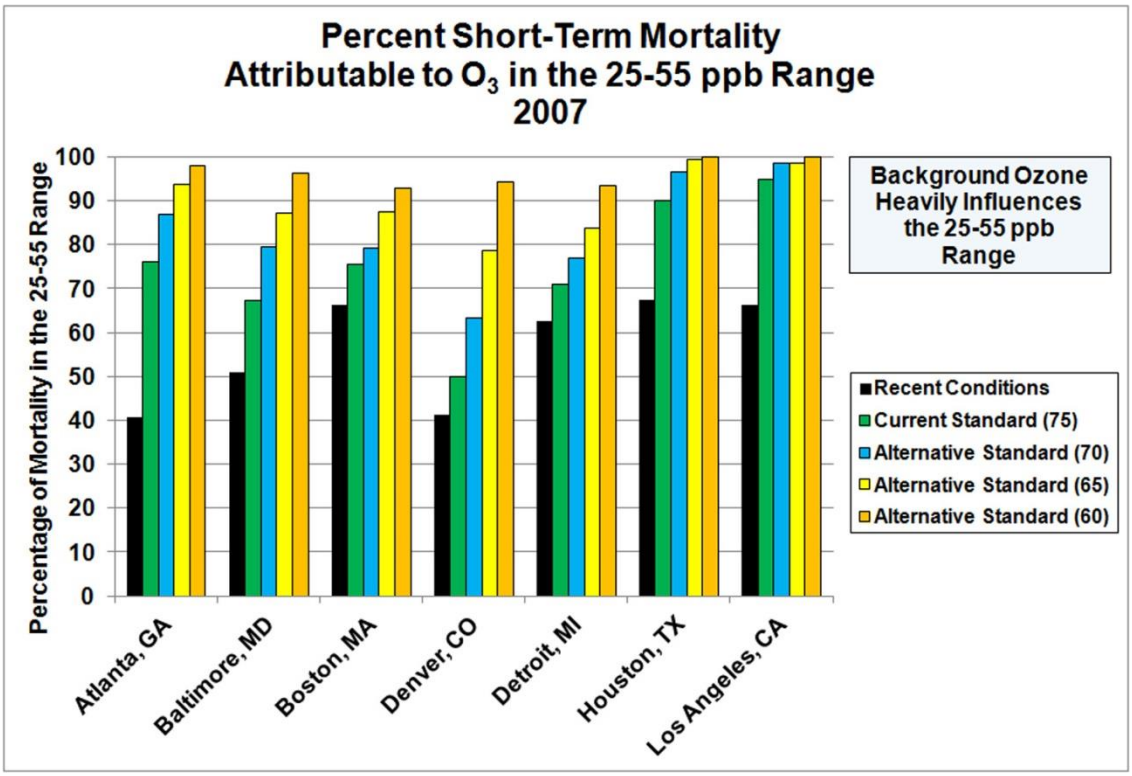


Figure 3-26. Percent short-term ozone-attributable mortality in the 25-55 ppb range for various exposure conditions for 2007 for 7 of 12 cities. (Source: Data from Fig. 7-B1 on page 7-B3 of EPA, 2014d).

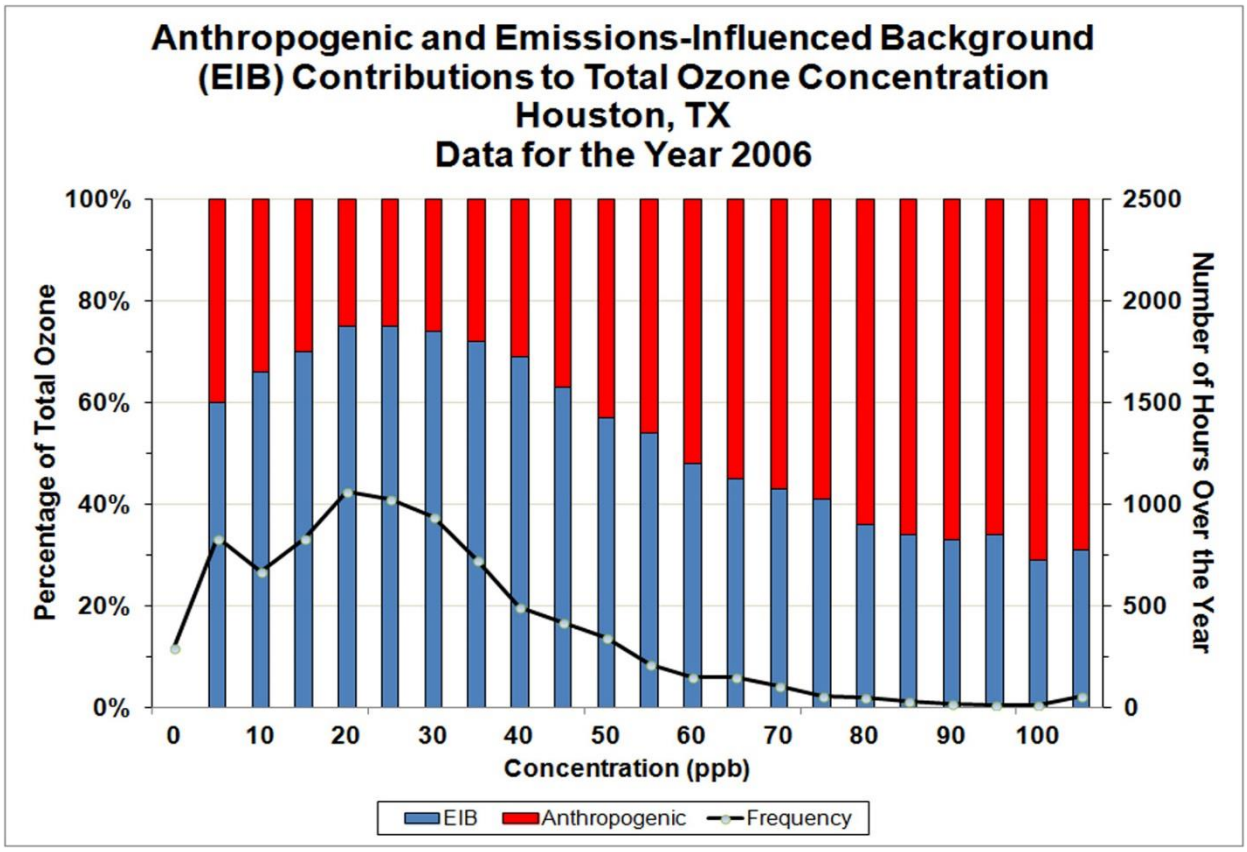


Figure 3-27. Binned (5 ppb) frequency distribution of observed hourly total O_3 (black curve; right axis) and average relative binned contributions of hourly maximum Emissions Influenced Background (EIB) and anthropogenic O_3 (bars; left axis) for Houston, TX (AQS ID 482010055) in 2006. Lefohn et al. (2014).

3.2.2 How is the Term *Background O_3* Defined?

From the previous section, it appears that the EPA desires to answer the question **"How much of the current ozone can be attributed to sources other than U.S. anthropogenic sources?** While the Agency is focused on how much of the current O_3 can be attributed to sources other than U.S. anthropogenic sources on days when ambient levels exceed the standard, as indicated above, as discussed previously, quantifying background O_3 is very important for assessing human health and ecological effects.

While background O_3 cannot be measured directly, estimating it accurately by utilizing both empirical and modeling approaches is important. Under a variety of meteorological conditions, background O_3 can make a substantial contribution to levels that result in exceedances of Federal standards when (1) ***episodic events*** occur under relatively clean conditions (Zhang et al., 2011; Lin et al., 2012a; Emery et al., 2012; Lefohn et al., 2014; Dolwick et al., 2015; Jaffe et al., 2018) and (2) local photochemical production combines with background levels that ***enhance*** the ambient concentrations (Lefohn et al., 2014; Dolwick et al, 2015). Background O_3 is of interest because (1) at times background O_3 is associated with high

concentrations experienced in the US Intermountain West that affect attainability of O_3 air quality standards (Lefohn et al., 2001; Langford et al., 2009; McDonald-Buller et al., 2011; Lin et al., 2012a; Dolwick et al., 2015) and (2) background O_3 contributes on a continuous basis to observed concentrations that influence human health and vegetation risk estimates, whose values influence recommended levels for Federal O_3 standards (McDonald-Buller et al., 2011; EPA, 2014a, 2014b). While considerable discussion in the US has focused on background contributions to the human health Federal O_3 primary standard, elevated background concentrations associated with stratospheric intrusions can affect vegetation (Skelly, 2000; FLAG, 2010).

The term "background O_3" in the United States over the years has not been defined consistently (McDonald-Buller et al., 2011; Lefohn et al., 2014; EPA, 2014a). EPA (2006) defined North American background (NAB) O_3 (previously referred to as Policy-Relevant Background by the US EPA) to include contributions from global anthropogenic and natural sources in the absence of North American (i.e., US, Canada, and Mexico) anthropogenic emissions. NAB O_3 is the range of concentrations that an air quality model estimates would exist in the absence of North American anthropogenic emissions. In 2013, the US EPA (2013) modified its definition of background by introducing the terminology US background (USB) O_3 concentrations. The level of USB O_3 is defined to include contributions from global anthropogenic and natural sources in the absence of US anthropogenic emissions. In other words, the USB ozone concentration is defined as the ozone concentration that would occur ***if all U.S. anthropogenic ozone precursor emissions were removed*** (draft ISA, Page ES-3). The difference associated with the hypothetical estimates using models of NAB O_3 and USB O_3 is small (EPA, 2013).

Recognizing that NAB O_3 and USB O_3 are estimated background concentrations that reflect hypothetical ***zero*** anthropogenic emissions, Lefohn et al. (2014) explored a new metric that estimated background O_3 levels under ***current anthropogenic emission*** conditions. The authors, using source-apportionment based modeling, referred to these background estimates as Emissions-Influenced-Background (EIB) O_3. The authors defined Emission-influenced Background (EIB) O_3 to include contributions from natural sources throughout the globe and from anthropogenic sources outside of North America. EIB O_3 estimates the impact of background sources, even in situations in which local O_3 has been influenced by US anthropogenic emissions. In August 2014 in its PA, the EPA (2014a) described estimates of source-apportionment US Background (USB_{AB}). The EPA (2014a) defined source-apportionment based US Background (USB_{AB}) in a similar manner as EIB O_3, except that USB_{AB} O_3 includes anthropogenic sources from Canada and Mexico. Similar to EIB, USB_{AB} estimates the impact of background sources, even in situations in which local O_3 has been influenced by US anthropogenic emissions (see Dolwick et al., 2015 for further clarification).

An important advantage in estimating either EIB O_3 or USB_{AB} background is that policymakers have an indication of (1) the relationship between current daily background levels and daily observed O_3 concentrations and (2) the level of O_3 concentration that may occur as a result of implementing emissions reductions strategies. For example, if EIB O_3 or USB_{AB} O_3 concentrations have a large relative contribution to observed O_3 concentrations at a specific

location, one would anticipate that emissions reductions on a regional scale would not have much impact on the concentrations at that site.

In the previous PA (EPA, 2014a), research results based on natural background were presented. Natural background O_3 is defined as the ozone concentrations that would occur if all anthropogenic emissions were removed worldwide. Processes that contribute to natural background O_3 include O_3 transport from the stratosphere and O_3 formed from precursor emissions originating from wildfires, lightning, natural methane sources, plants, and other natural VOC and NO_X emissions.

On page 1-6 of the draft ISA, the authors mention ***Baseline ozone*** as an alternative metric for USB and NAB. Baseline O_3 has been defined as the measured O_3 concentration at rural or remote sites that have not been influenced by recent, local emissions (Jaffe et al., 2018). In contrast to USB, baseline O_3 is directly measured. Baseline measurements are typically from monitors in locations that are minimally influenced by local anthropogenic sources, and samples used as baseline measurements are limited to those monitored during meteorological conditions consistent with the relative absence of local contamination. Baseline O_3 can include the O_3 produced from U.S. emissions that circle the globe and may also include effects of same-state emissions. An example of the latter would be ozone from U.S. emissions near the West Coast or Gulf Coast that is transported over the Pacific Ocean or Gulf of Mexico, respectively, and then transported back onshore. In some cases, sources that impact baseline O_3 may not similarly impact O_3 in populated locations. For instance, baseline O_3 measured on a mountaintop may include stratospheric influences that are not representative of contributions in nearby lower elevation locations.

The draft ISA points out (Page 1-7) that there are several reasons why baseline O_3 measurements cannot be used as a proxy to estimate USB O_3 levels in urban areas. As previously described, baseline O_3 can include contributions from U.S. emissions. Additionally, baseline O_3 monitors can be very distant from urban sites, and O_3 measured at the baseline site can be destroyed through surface deposition or chemical reactions during transport from the baseline site to a downwind monitor. In addition, atmospheric conditions may not favor transport of baseline O_3 from the monitor location to populated areas at lower elevations. The draft ISA also points out that another reason why baseline O_3 measurements cannot be used as a proxy for USB O_3 levels (using the zero-out methodology) in urban areas is that meteorological conditions that favor mixing from the free troposphere to ground level have strong ventilation and are not conducive to photochemical O_3 episodes that produce the highest urban O_3 concentrations. However, as noted in the ISA, stratospheric intrusion events are an exception. The draft ISA concludes (Page 1-7) that while baseline O_3 measurements cannot be used directly to estimate USB (zero-out methodology) O_3, baseline O_3 data are useful for evaluating the CTMs that are used to provide model estimates of USB (zero-out) O_3.

In summary, the following terms have been used in the draft ISA to describe background O_3:

- USB is defined to include contributions from global anthropogenic and natural sources in the absence of US anthropogenic emissions.

- NAB has been defined as the O_3 concentration that would occur in the U.S. in the absence of anthropogenic emissions in continental North America (U.S. EPA, 2013). NAB has also been referred to as policy-relevant background (PRB) in earlier publications (U.S. EPA, 2007).

- Emissions-influenced background (EIB) has been defined as another measure of background O_3 estimated from source apportionment modeling approaches while including chemical interactions with anthropogenic emissions (Lefohn et al., 2014).

- Source-apportionment US Background (USB_{AB}) is the amount of O_3 formed from sources other than U.S. anthropogenic sources as estimated via an apportionment technique (Dolwick et al., 2015). USB_{AB} O_3 includes anthropogenic sources from Canada and Mexico.

- Natural background O_3 is defined as the O_3 concentrations that would occur if all anthropogenic emissions were removed worldwide. Processes that contribute to natural background O_3 include O_3 transport from the stratosphere and O_3 formed from precursor emissions originating from wildfires, lightning, natural methane sources, plants, and other natural VOC and NO_X emissions.

- Baseline O_3 has been defined as the measured O_3 concentration at rural or remote sites that have not been influenced by recent, local emissions (Jaffe et al., 2018). The draft ISA points out (Page 1-7) that there are several reasons why baseline O_3 measurements cannot be used as a proxy to estimate USB O_3 levels in urban areas.

USB, as well as USB_{AB}, is a model construct that cannot be measured using ambient monitoring data. The draft ISA (EPA, 2019a) notes that this approach is consistent with the 2006 Ozone Air Quality Criteria Document (AQCD) (U.S. EPA, 2006a) and the 2013 Ozone ISA (U.S. EPA, 2013), which also used modeled estimates of background O_3. Reliance on atmospheric modeling for USB, as well as USB_{AB} concentrations estimates, continued in the 2013 Ozone ISA (U.S. EPA, 2013). In earlier assessments, O_3 estimates were based on measurements at monitoring sites with low concentrations that appeared to be isolated from anthropogenic sources (Altshuller and Lefohn, 1996; Trainer et al., 1993).

3.2.3 EPA's Preference for the Use of USB Rather than Other Definitions of Background

EPA has preferred to use USB methodology for estimating background O_3. No clear reason is provided why the authors of the draft ISA preferred the USB (i.e., zero-out) approach rather than other modeling methodologies, for characterizing background O_3 in the document. On page 1-4, the draft ISA notes:

> ***In this document, the term U.S. background (USB) is used to assess background ozone*** emphasis added). The USB concentration is defined as

> the ozone concentration that would occur if all U.S. anthropogenic ozone precursor emissions were removed.

The authors further note on page 2-34 of the draft PA:

> The methodologies reviewed range in complexity from simply turning off U.S. anthropogenic (or specific sources) emissions, to normalizing derivatives from instrumented models, to complex tagging techniques (e.g., CAMx OSAT, APCA, or Grewe, 2013).[27] This analysis follows the zero-out approach for simplicity of interpretation and consistency with previous analyses.

On page 2-59 of the draft PA, the authors state:

> The overall findings of this assessment are consistent with the 2014 O_3 PA, with the EPA's Background Ozone whitepaper (U.S. EPA, 2015), and with the peer reviewed literature.

In the EPA White Paper (EPA, 2015) that is referred to in the draft PA, the Agency noted that

> For the purposes of this white paper and the continuing discussion of background O_3 issues in the NAAQS implementation context, the EPA considers background O_3 to be any O_3 formed from sources or processes other than U.S. manmade emissions of nitrogen oxides (NOx), volatile organic compounds (VOC), methane (CH4), and carbon monoxide (CO).7 This definition of background is specifically referred to as U.S. background (USB).8

While the EPA in its White Paper (EPA, 2015) apparently made a unilateral decision to use USB rather than USB_{AB} in defining the term "background O_3", no clear rationale was apparently provided.

On page 1-5 of the draft ISA, the EPA notes that modeling approaches for estimating background O_3 can be classified as either source-sensitivity or source-apportionment approaches. USB was originally estimated using source-sensitivity approaches. Apportionment-based USB (USB_{AB}) has been defined as the amount of ozone formed from sources other than U.S. anthropogenic sources as estimated via an apportionment technique (Dolwick et al., 2015). In the 2014 Policy Assessment (EPA, 2014), the Agency discussed both USB and USB_{AB}.

The draft ISA notes on page 1-52:

> The zero-out approach is more suited for answering the question "what ozone levels would exist in the absence of all U.S. emissions?" while the source apportionment approach is more suited for answering the question "what amount of current ozone comes from background sources?" The

> difference between USB and USB_{AB} is small in remote areas most strongly affected by USB sources, but can be substantial in urban areas strongly affected by anthropogenic sources that influence both production and destruction of ozone (Dolwick et al., 2015).

Given that the EPA has chosen to use USB rather than USB_{AB}, it must be remembered that USB estimates will represent a quantity never to occur in the real atmosphere (EPA, 2014a). As noted in the 2014 PA document (EPA, 2014a), sensitivity approaches (i.e., USB) can be unreliable for evaluating mass contributions to O_3 production because of nonlinearity in the chemistry.

The US EPA (2014a) noted that the strength of the source-apportionment approach (i.e., USB_{AB}) is that it provided a direct estimate of the amount of O_3 contributed by each source category, while avoiding artifacts caused by non-linearity in the chemistry, which is a potential with the zero emissions (i.e., zero-out) modeling used to estimate USB O_3 concentrations. Table 3-1 below (original labeled Table 2-1 on page 2-15 in EPA, 2014a) is reproduced from the EPA's Policy Assessment document (EPA, 2014a). The table compares the two model methodologies used to characterize USB (i.e., zero-out) and USB_{AB} (apportionment based).

As noted in the draft ISA (EPA, 2019a) (Page IS-15):

> Both approaches are essential and complementary for understanding and estimating USB ozone. The zero out approach is suited for determining what ozone levels would have existed in recent modeled years in the absence of all U.S. emissions, while the source apportionment approach is suited for determining the fraction of current ozone originating from background sources in recent modeled years.

As noted above, a key point made in the draft ISA (page 1-52) is that the difference between USB and USB_{AB} is small in remote areas most strongly affected by USB sources, ***but can be substantial in urban areas strongly affected by anthropogenic sources that influence both production and destruction of ozone*** (Dolwick et al., 2015).

Estimation Methodology	Question addressed	Background Quantities	Strengths and Limitations
Zero-out	How much ozone would remain if controllable emissions were completely removed?	NB / NAB / USB	Strength: The approach is simple to implement and provides an estimate of the lowest O_3 levels that can be attained by eliminating all U.S. anthropogenic emissions. Limitation: Estimates are based on a counterfactual, represents a quantity never to occur in real atmosphere. Additionally, sensitivity approaches can be unreliable for evaluating mass contributions to O_3 production because of non-linearity in the chemistry.
Source Apportionment	How much of the current ozone can be attributed to sources other than U.S. anthropogenic sources?	Apportionment-based USB	Strength: Provides a direct estimate of the amount of O_3 contributed by each source category while avoiding artifacts caused by non-linearity in the chemistry. Limitation: While this approach identifies important sources that contribute to O_3, it does not predict quantitatively how O_3 will respond to specific emissions reduction scenarios.

Table 2-1 Comparison of the two model methodologies used to characterize background ozone levels.

Table 3-1. A comparison of the two model methodologies used to characterize background ozone levels. Source: Source: US EPA (2014a).

The distinction between USB and USB_{AB} is important because apportionment techniques for estimating USB_{AB} are designed to realistically treat nonlinear and nonadditive interactions of USB and U.S. anthropogenic emissions that affect both production and destruction of O_3. In contrast, source-sensitivity modeling approaches originally used for estimating USB are not designed to address these interactions. As pointed out in the draft ISA (page 1-5), USB and USB_{AB} are **not** the same quantity estimated with different approaches but are ***estimates of***

conceptually different quantities*. *While USB is an estimate of ozone concentrations that could be achieved if U.S. anthropogenic sources were eliminated, USB_{AB} is an estimate of how much ozone can be attributed to background sources when those anthropogenic sources are present.

As pointed out in Table 3-1 above (Table 2-1 in the 2014 PA), the USB approach is simple to implement and provides an estimate of the lowest O_3 levels that can be attained by eliminating all US anthropogenic emissions. The table also notes that the USB estimates are based on a counterfactual, represents a quantity never to occur in the real atmosphere. As noted in the 2014 PA document (EPA, 2014a), sensitivity approaches can be unreliable for evaluating mass contributions to O_3 production because of nonlinearity in the chemistry. The USB_{AB} approach provides a direct estimate of the amount of O_3 contributed by each source category, while avoiding artifacts caused by non-linearity in the chemistry. As noted in the 2014 PA (EPA, 2014a), while the approach identifies important sources that contribute to O_3, it does not predict quantitatively how O_3 will respond to specific emissions reductions scenarios. However, it is reasonable to assume that as emissions were reduced with the result that current O_3 levels at the high end were reduced and the lower levels shifted toward the mean due to reduction in NO_x levels, the USB_{AB} would ***increase*** to levels above the current USB_{AB} estimates based on current O_3 levels. *In other words, background O_3 would make up a higher fraction of the levels observed in future O_3 levels achieved with emission reductions*.

As pointed out in Table 3-1 (Table 2-1 in the 2014 PA), it appears that USB_{AB} should be used rather than USB to answer the question "How much of the current ozone can be attributed to sources other than U.S. anthropogenic sources?"

A quantitative understanding of background O_3 is essential for air quality management. This is especially true given the recent lowering of the NAAQS O_3 levels and the associated increasing relative importance of background O_3 as domestic precursor emissions decrease.

Thus, we are left with the dilemma of why the EPA estimated USB rather than USB_{AB} for better clarifying background O_3 levels. Simply stating that its analysis follows the zero-out approach for simplicity of interpretation and consistency with previous analyses is not a strong rationale. In the previous NAAQS O_3 rulemaking, the Policy Assessment document in 2014 (EPA, 2014a) included discussion of USB_{AB} estimates of background O_3 because of the importance of the estimates in answering the regulatory questions. The EPA estimated 2007 seasonal (i.e., April through October) mean USB MDA8 O_3 concentrations using a combination of the GEOS-Chem global model and the Community Multi-scale Air Quality (CMAQ) (zero out) and CAMx (source apportionment) regional models. Dolwick et al. (2015) summarized for the western US both the USB and USB_{AB} findings contained within the Policy Assessment document (EPA, 2014a). EPA in the 2014 PA estimated USB_{AB} levels for understanding and estimating background O_3 for determining the fraction of current O_3 originating from background sources in recent modeled years. The use of only background O_3 estimates associated with USB (i.e., the zero-out modeling and its non-linearity chemistry problems) may result in more inconsistency than would have occurred if USB_{AB} had been estimated. An important question is: Even if inconsistencies exist, are these inconsistencies important when assessing (1) the fraction of background O_3 when concentrations are highest under current

ambient conditions and (2) the role of background O_3 for quantifying human health and vegetation risk?

3.2.4 EPA's Conclusion on Background O_3 Modeling Estimates

EPA (draft ISA, page 1-56) notes that while the seasonal mean USB concentration patterns may be important for identifying atmospheric processes leading to high USB concentrations, ***they are less relevant for estimating USB concentrations on days with high MDA8 concentrations and for understanding the role that hourly average background O_3 concentrations play in affecting human health and vegetation risk estimates.*** On page 1-5 of the draft ISA, the EPA notes that the averaging time of a USB estimate is intended to match the averaging time of the total O_3 concentration measured. In other words, if the EPA desired to determine the percentage of background O_3 associated with the top ten daily maximum 8-h concentrations during the year at a specific monitoring site, the background O_3 must be reported in daily maximum 8-h concentrations.

Focusing on the seasonal means, whose values may be of interest to scientists attempting to understand atmospheric processes, on Page 1-55 in the draft ISA (EPA, 2019a), the EPA notes that the 2013 Ozone ISA (U.S. EPA, 2013) reported higher mean USB and NAB concentration estimates in spring than in summer for most regions of the U.S, and these results are consistent with earlier modeling estimates. However, EPA notes that some new results are consistent with this pattern (e.g., Lefohn et al., 2014), while other results suggest that summer USB and baseline O_3 concentrations can be comparable to (Jaffe et al., 2018) or greater than (Guo et al., 2018) spring concentrations. Guo et al. (2018) reported region-wide seasonal mean USB concentrations greater in summer than spring for most U.S. regions. The authors proposed that improvement of isoprene-NO_X chemistry was the reason for the difference in results compared to previous results that indicated springtime was the period of greatest background O_3 contributions.

EPA in the draft ISA (page 1-55) believes this to be significant because numerous studies of USB and other measures of background O_3 have focused on spring as the season with the greatest USB concentrations. The draft ISA (page 1-55) notes that

1. Recent publications have come up with conflicting conclusions about seasonal trends in USB. Higher seasonal mean USB concentrations in spring than in winter were reported for intermountain western sites (Fiore et al., 2014).

2. Fiore et al. (2014) reported higher seasonal mean NAB concentrations in spring than in summer at high-elevation western U.S. sites, consistent with the 2013 Ozone ISA (EPA, 2013).

3. Region-wide seasonal mean USB concentrations greater in summer than spring were reported for most U.S. regions (Guo et al., 2018). Improvement of isoprene-NO_X chemistry was proposed as the reason for the difference in results compared with earlier modeling results like those of (Fiore et al., 2014).

4. Jaffe et al. (2018) reported comparable median spring and summer baseline ozone concentrations at elevations >1 km in the western U.S., while below 1-km baseline ozone concentrations were higher in spring.

The authors of the draft ISA did not resolve the conflicting conclusions about when seasonal mean background O_3 is greatest. I suggest that further discussion is warranted concerning the inconsistencies in the results. Currently, the draft ISA appears to dismiss the inconsistencies by indicating that seasonal mean USB metrics are less relevant for estimating USB concentrations when focusing on days with high MDA8 concentrations, as well as for understanding the role that hourly average background O_3 concentrations play in affecting human health and vegetation risk estimates.

On page 17 of Jaffe et al. (2018), the authors note that model-calculated USB O_3 was greatest in March through June, which agrees with the observations when stratospheric contributions are greatest at many high- and low-elevation sites across the U.S. (Lefohn et al., 2011, 2012). Jaffe et al. (2018) summarized their findings as follows concerning the seasonal behavior of background O_3:

> Model-calculated USB O_3 is greatest in March through June, with monthly mean MDA8 mole fractions at higher elevations in the west of up to 50 ppb and annual 4th highest MDA8 values exceeding 60 ppb at some locations. Lower elevation cities nationwide have monthly mean USB O_3 of 20–40 ppb during the O_3 season. Daily variations, particularly in spring and early summer, can be due to stratospheric intrusions mixed with Asian pollution, which can contribute to observed MDA8 values over 70 ppb.

Using baseline O_3 data (Fig. 2 of Jaffe et al., 2018), Jaffe et al. (2018) illustrate the vertical profiles of O_3 at 4 sites in the West (Trinidad Head, Cheeka Peak, Mt. Bachelor Observatory, and Chews Ridge). The authors note that at low elevations at the four sites, mean spring O_3 levels are about 10 ppb higher than summer values, whereas above 1 km, median spring and summer values are comparable with summer.

Guo et al. (2018) note that their model may exaggerate the relative importance of enhanced background O_3 resulting from soil NOx and isoprene. The authors noted in their paper that substantial biases in the severity and timing of high-O_3 events occurred in their model and that the model underestimated the frequency of high events in spring that they indicated were possibly associated with stratospheric intrusions. These important uncertainty statements mentioned in their paper may help explain their finding that USB O_3 tended to be higher in the summer than in the spring in most regions.

I believe that there continues to be strong evidence as supported in the literature that background O_3 across the US is highest at many sites during the springtime (including into the month of June) and is an important contributor at many high-elevation sites throughout the year. I would suggest that the authors of the draft ISA evaluate O_3 data in the EPA's AQS database and quantify when the highest O_3 exposures occur at various types of sites. This I believe would add to the discussion. As noted earlier (Section 3.1.2), actual O_3 monitoring data show that the

highest O_3 exposures for the National Park sites occur at some sites across the US during the springtime (March to mid-June). The EPA in its 2014 PA (EPA, 2014c) (Welfare Appendix, page 7A-12) provided the highest 3-month W126 values and the timeframe corresponding to those W126 exposures for the National Parks with O_3 monitors for the period 2006-2010. Using hourly average O_3 data from 57 National Parks, Table 7A-2 was provided earlier. Note that several of the O_3 monitors in the National Parks experienced their highest W126 exposures during the spring months (defined as March, April, May or April, May, June) period. While the months of April, May, and June are not entirely a spring period (half of June is still spring), the time of year when the frequency of stratospheric intrusions (i.e., a natural process) to the surface is greatest at many sites during the March – June window. Besides the National Parks, there are other monitoring data with the EPA's AQS database where the highest O_3 exposures occur during the March-June period.

Earlier we had discussed why background O_3 is important. While seasonal mean background O_3 estimates are of interest for identifying atmospheric processes, there use in assessing the role of background O_3 on exceedance days, as well as in human health and vegetation risk analyses, is limited. The time series of 8-h average daily maximum concentrations of background O_3 is important for some human health risk analyses and hourly average background O_3 concentrations are important for vegetation risk analyses. It appears in the reading the draft ISA and draft PA that the EPA is focused on understanding the percent contribution of background O_3 to current ambient levels on days when ambient levels exceed the standard. However, as noted earlier, there are other considerations in the rulemaking process that are of equal importance to quantifying background O_3. In many cases, assessment of human health and ecological effects is based on understanding the role that background O_3 plays throughout the distribution of hourly average concentrations and not just the role that background O_3 plays during periods of highest O_3 exposures. For the vegetation related W126 exposure index, each hourly average concentration is weighted and accumulated over a specific period for assessing vegetation effects. Hourly average background O_3 concentrations contribute to the observed concentrations and therefore, contribute to the cumulative risk. For some human health risk assessments, daily 8-h average concentrations are used in a time series. Daily maximum 8-h average concentrations contain background O_3 concentrations, which contribute to the estimated human health risk.

3.2.5 How much of the current ozone can be attributed to sources other than U.S. anthropogenic sources?

To answer the question of how much of the current O_3 can be attributed to sources other than U.S. anthropogenic sources both USB_{AB} (EPA, 2014; Dolwick et al., 2015) and EIB (Lefohn et al., 2014) daily estimates are used for illustrative purposes. Besides focusing on the highest hourly average O_3 concentrations (i.e., the upper end of the distribution of hourly values), it is important to understand the relative role that background O_3 plays over the entire distribution of total ambient O_3 concentrations. For example, for the human health risk associated with epidemiological models using no cutoff (i.e., threshold), the lower end of the distribution plays an important role. Background O_3 contributes a large amount to the lower

concentrations and cannot be reduced with reductions in emissions. For example, the high-elevation Yellowstone National Park site in Wyoming is dominated by background O_3 throughout the year with minor anthropogenic contributions (Lefohn et al., 2014). In Figure 3-28 below, the relative comparison of EIB background levels (noted by blue) to anthropogenic (noted by red) within each concentration level shows that background contributes greater than 80%, including the mid-range concentrations (20-25 ppb), which was an important range that influenced EPA's human health risk estimates in 2014. As noted above, EIB O_3 for 2006 is similar to the USB_{AB} estimates for 2007 utilized by the EPA (2014) in its Policy Assessment document in 2014 (EPA, 2014a). In comparison, Figure 3-29 illustrates for Denver the contribution of background within the mid-range concentrations is approximately 75 to 80%. For the Los Angeles area (Figure 3-30), a site heavily influenced by anthropogenic emissions, background contributes 60-80% in the mid-range.

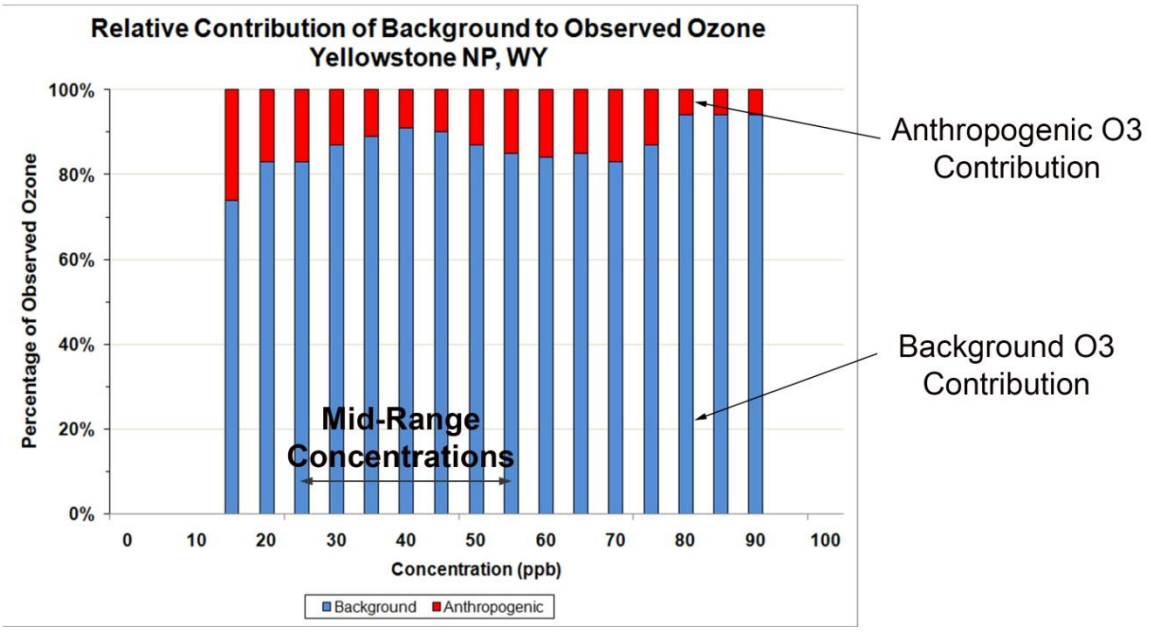


Figure 3-28. Average relative contributions of current hourly background (blue) and anthropogenic ozone (red) for Yellowstone NP (WY) (AQS ID 560391011) in 2006. (Source: Lefohn et al., 2014).

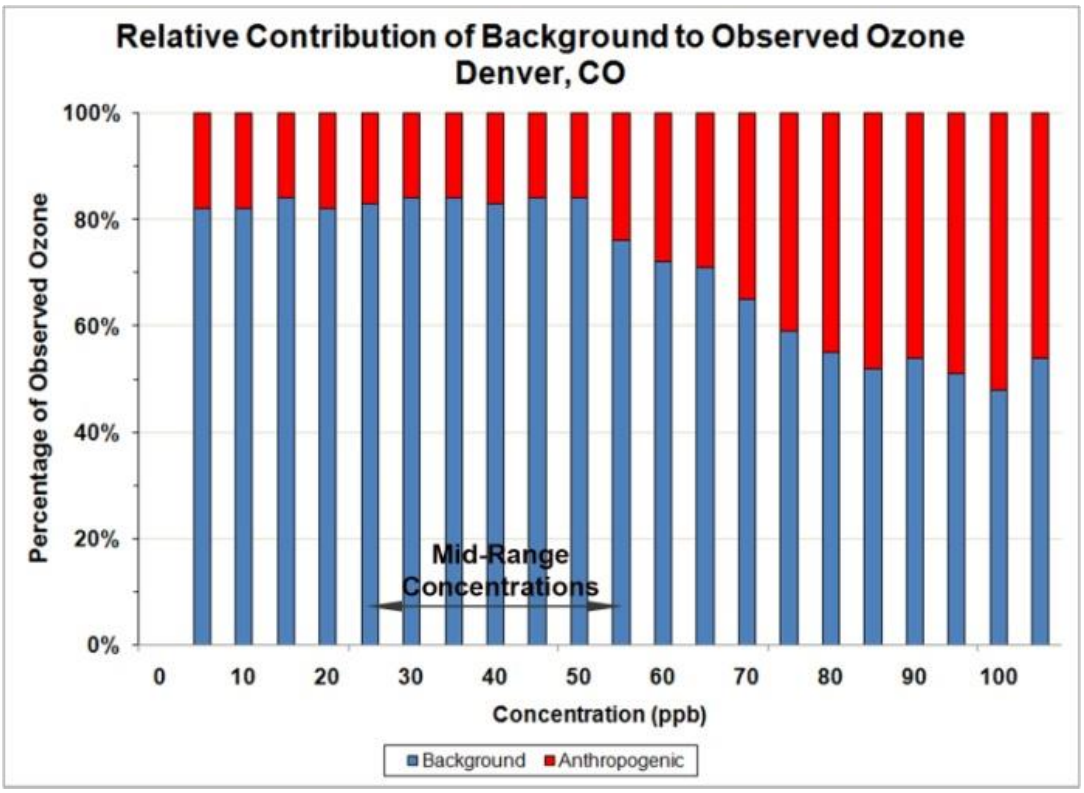


Figure 3-29. Average relative contributions of current hourly background (blue) and anthropogenic ozone (red) for Denver (CO) (AQS ID 080590006) in 2006. (Source: Lefohn et al., 2014).

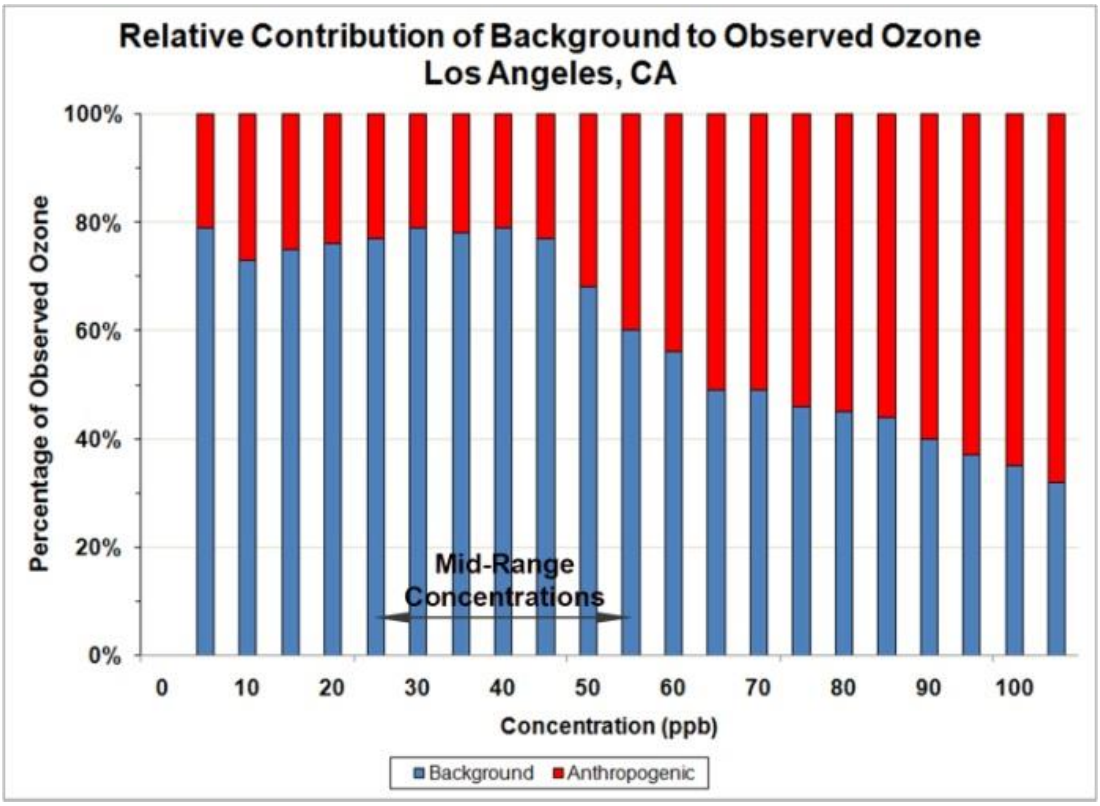


Figure 3-30. Average relative contributions of current hourly background (blue) and anthropogenic ozone (red) for Los Angeles (CA) area (AQS ID 060719004) in 2006. (Source: Lefohn et al., 2014).

On page 2-23 of the EPA's Policy Assessment document (EPA, 2014a), Fig. 3-31 below (Fig. 2-15 in EPA, 2014a) illustrates the distributions of the relative proportion of apportionment-based U.S. Background (USB_{AB}) to total O_3 (all site-days), binned by modeled MDA8 from the 2007 source apportionment simulation. The figure indicates that the USB_{AB} concentrations play a very important role in both the low and mid-range total O_3 concentrations (EPA, 2014a).

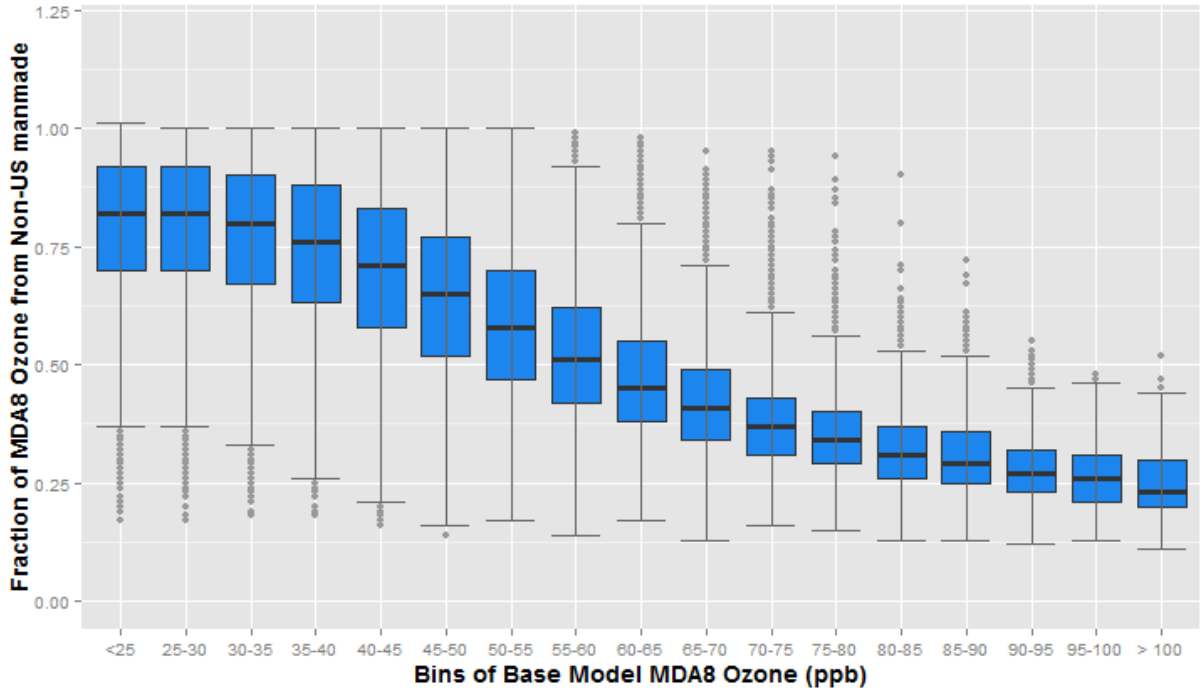
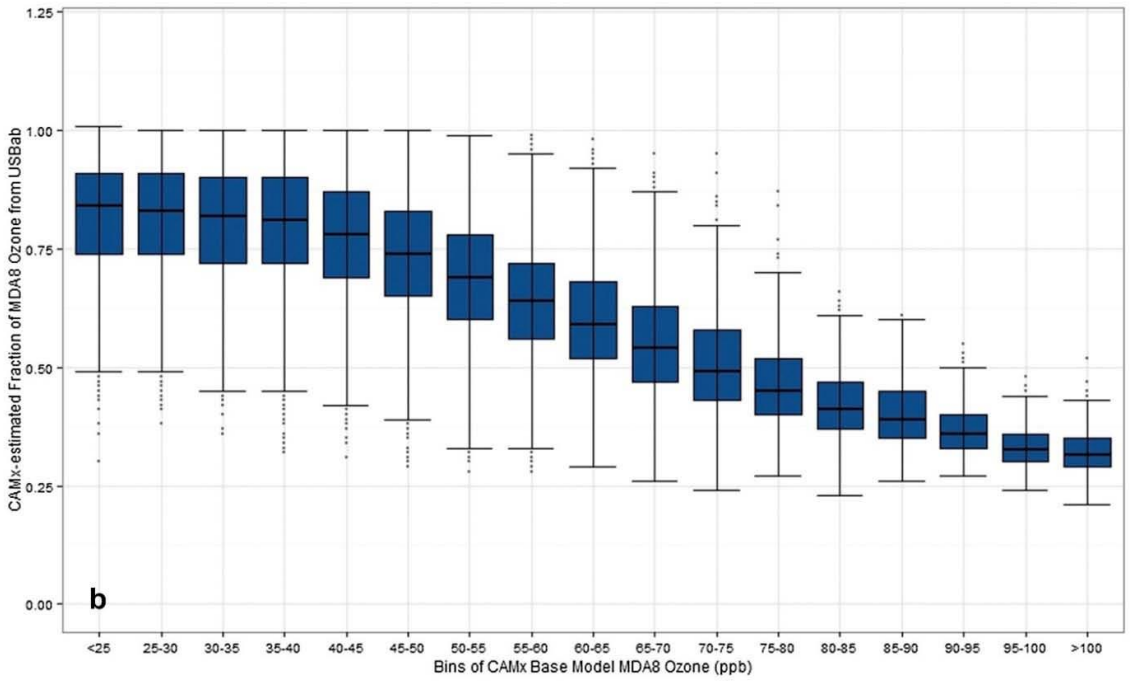


Figure 3-31. Distributions of the relative proportion of apportionment-based U.S. Background (USB_{AB}) to total O_3 (all site-days), binned by modeled MDA8 from the 2007 source apportionment simulation. Source: Fig. 2-15 in EPA (2014a) with slight modification.

In Fig. 3-32 below (Fig. 1-14b on Page 1-59 of draft ISA) illustrates CAMx estimates of daily distributions of bias-adjusted USB_{AB} O_3 fraction at monitoring locations across the western U.S. for the period April−October 2007, binned by base model MDA8 ozone concentration ranges. Similar to the results presented in EPA's Policy Assessment document (EPA, 2014a), the USB_{AB} concentrations play a very important role in both the low and mid-range total O_3 concentrations.



ppb = parts per billion.
Source: Dolwick et al. (2015).

Figure 3-32. CAMx estimates of daily distributions of bias-adjusted apportioned-based (USB_{AB}) ozone fraction at monitoring locations across the western U.S. for the period April−October 2007, binned by base model MDA8 ozone concentration ranges. Source: Draft ISA – page 1-59 figure 1-14b).

On page 1-1 of the draft ISA, the authors note that

> U.S. background ozone continues to account for a large fraction of ambient ozone concentrations as a result of stratospheric exchange, international transport, wildfires, lightning, global methane emissions, and natural biogenic and geogenic precursor emissions. New results concerning U.S. background ozone are (1) a wider range of concentration estimates, and poorer agreement between models have been observed than were reported in the 2013 Ozone ISA, with a range of uncertainty of ~10 ppb for seasonal average concentrations, (2) U.S. background concentrations are uncorrelated with local ground-level concentrations above ~60 ppb, and (3) an increasing trend of U.S. background concentration at high elevation western U.S. sites before approximately 2010 now shows signs of slowing or even reversing, ***probably*** (emphasis added) due to decreasing East Asian precursor emissions.

Item (2) above indicates that background O_3 concentrations do not increase with total O_3 concentration above 60 ppb and according to Dolwick et al. (2015) this results in decreasing predicted relative contributions of background O_3 to total O_3 at the higher total O_3 concentrations (page 1-56). The draft ISA pointed out (page 1-56) that Lefohn et al. (2014) also described a

decreasing trend of relative background (identified by the authors as "Emissions Influenced Background") contribution with increasing total O_3 concentration. At low-elevation and urban sites in the western U.S., O_3 concentrations estimated as USB, USB_{AB}, NAB, or EIB contributions were also reported to be independent of overall O_3 concentration, resulting in a decreasing relative background contribution with increasing total O_3 concentration (Guo et al., 2018a; Guo et al., 2018b; Dolwick et al., 2015; Lefohn et al., 2014). However, model results do show increasing USB_{AB} and NAB concentrations with increasing O_3 concentration at high-elevation western U.S. sites (Fiore et al., 2014; Lefohn et al., 2014).

3.2.6 Sources of Background Ozone

On page 1-1 of the draft ISA, the authors conclude that U.S. background O_3 continues to account for a large fraction of ambient O_3 concentrations as a result of stratospheric exchange, international transport, wildfires, lightning, global methane emissions, and natural biogenic and geogenic precursor emissions. At noted in the draft ISA, as the literature on background O_3 has evolved, much of the discussion has focused on the relative importance of stratospheric O_3 and intercontinental transport as the major sources of background O_3 (Page IS-13 of the draft ISA).

On Page IS-13 of the draft ISA, the authors note that tropospheric O_3 derived from stratosphere-troposphere dynamics was described in detail in the 2013 Ozone ISA (U.S. EPA, 2013). Stratospheric air naturally rich in O_3 can be transported into the troposphere under certain meteorological circumstances, with maximum contributions observed at midlatitudes during the late winter and early spring. This process, known as "tropopause folding," is characterized by episodic events typically lasting a few days from late winter through spring when deep stratospheric intrusions rich in O_3 can quickly and directly well into the troposphere and, more rarely, reach ground level (U.S. EPA, 2013). The 2013 Ozone ISA (U.S. EPA, 2013) also discussed the potential importance of deep convection, another form of stratosphere-troposphere exchange that occurs mainly in summer, as a mechanism for transporting stratospheric O_3 into the upper troposphere. Stratospheric intrusion events related to frontal passage and tropopause folding that reach the surface have less influence on surface O_3 during the summer months.

The relevance of stratospheric-to-tropospheric transport (STT) for influencing low-tropospheric O_3 concentrations has been well documented (e.g., Reed, 1955; Junge, 1962; Danielsen, 1968; Danielsen, 1974; Danielsen and Mohnen, 1977; Ludwig et al., 1977; Shapiro, 1980; Haagenson et al., 1981; Davies and Schuepbach, 1994; Lamarque and Hess, 1994; Schuepbach et al., 1999; Stohl et al., 2000; Lefohn et al., 2001; Cooper et al., 2005; Cristofanelli et al., 2006; Hocking et al., 2007; Ordóñez et al., 2007; Langford et al., 2009; Akriditis et al., 2010; Cristofanelli et al., 2010; Škerlak et al. (2014, 2019). Lefohn et al. (2001) (authors A.S. Lefohn, S.J. Oltmans, T. Dann, and H.B. Singh) described the contribution of stratospheric O_3 to observed O_3 levels across the U.S. The authors attributed STT processes to the observation that hourly average O_3 concentrations $\geq$ 50 ppb occurred frequently during the photochemically quiescent months in the winter and spring at several rural sites across southern Canada and the northern US. In their paper, the authors described an STT event, where the stratosphere contributed on May 6, 1999 to enhanced O_3 concentrations in Boulder, Colorado. Data were provided courtesy of A. Langford of NOAA-Boulder. Eight years later, Langford et al. (2009)

discussed the May 6, 1999 contribution of stratosphere-to-troposphere transport to high surface O_3 along the Colorado Front Range using lidar and surface measurements. In their paper, the authors indicated that the magnitude of O_3 background was controversial and that Lefohn et al. (2001) described that frequent occurrences of 50–80 ppb O_3 at remote northern U.S. sites during springtime (1988 to 1998) implied that significantly higher background values occurred. Langford et al. (2009) noted that Lefohn et al. (2001) attributed these high O_3 values to a larger stratospheric source than was shown in models and suggested that it might be difficult to satisfy the NAAQS for O_3 in locations where the stratospheric influence was significant.

Langford et al. (2009) indicated that others disputed the Lefohn et al. (2001) findings and used results from models to argue that the high-O_3 episodes described by Lefohn et al. (2001) could be explained by subsidence of free tropospheric air contaminated by North American anthropogenic sources and thus, did not represent true background values. Langford et al. (2009) felt that the modeling results were at odds with many other studies, which have presented evidence for significant stratospheric contributions to surface O_3 at both high-altitude sites (Schuepbach et al., 1999; Stohl et al., 2000) and near sea level (Cooper et al., 2005; Hocking et al., 2007). Langford et al. (2009) showed additional examples of deep STT contributing to high surface O_3 using lidar and surface measurements from the Front Range of the Colorado Rocky Mountains during the 1999 O_3 season (March–October). Their results showed that the stratospheric source was not only significant but could directly lead to exceedances of the 2008 NAAQS standards in a major metropolitan area. Langford et al. (2009) described a deep tropopause fold brought ~215 ppb of O_3 to within 1 km of the highest peaks in the Rocky Mountains on 6 May 1999. One-minute average O_3 mixing ratios exceeding 100 ppb were subsequently measured at a surface site in Boulder, and daily maximum 8-hour O_3 concentrations greater or equal to the 2008 NAAQS O_3 standard of 0.075 ppm were recorded at 3 of 9 Front Range monitoring stations. Other springtime peaks in surface O_3 were also shown to coincide with passage of upper level troughs and dry stable layers aloft. The authors noted that their results showed that the stratospheric contribution to surface O_3 was significant and could lead to exceedance of the 2008 NAAQS O_3 standards in a major U.S. metropolitan area.

Lefohn et al. (2011), using trajectory calculations, investigated the frequency of STT events and their associated enhancements on 12 surface O_3 monitoring sites in the western and northern tier of the US. The trajectory model introduced by Wernli and Davies (1997) was used to identify days of high probability for STT trajectories to enhance surface O_3 at specific monitoring sites. For most of the sites analyzed, Lefohn et al. (2011) indicated that contributions from stratosphere-to-troposphere transport to the surface (STT-S) were frequent during specific months and appeared to enhance the surface O_3 concentrations at both high- and low-elevation monitoring. Lefohn et al. (2012), quantified the frequency of STT events that result in O_3 concentration enhancements (i.e., hourly average concentrations $\geq$ 50 ppb) observed at 39 high- and low-elevation monitoring sites in the US during the years 2007-2009. They employed a forward trajectory-based approach to address the relationship between stratospheric intrusions and enhancements in hourly average O_3 concentrations. Their results indicated that STT down to the surface (STT-S) frequently contributed to enhanced surface O_3 hourly averaged concentrations at sites across the US, with substantial year-to-year variability. The O_3 concentrations associated with the STT-S events appeared to be large enough to enhance the measured O_3 concentrations during specific months of the year. Months with a statistically

significant coincidence between enhanced O_3 concentrations and STT-S occurred most frequently at the high-elevation sites in the Intermountain West, as well as at the high-elevation sites in the West and East. These sites exhibited a preference for coincidences during the springtime and in some cases, the summer, fall, and late winter. Besides the high-elevation monitoring sites, low-elevation monitoring sites across the entire US experienced enhanced O_3 concentrations coincident with STT-S events. Škerlak et al. (2014) noted that STT processes, which contribute to background O_3, affect the Intermountain West and other mountain ranges in the West year around, with a clear peak during the spring.

The STT-S "hits" as has been estimated using the methodology described in Lefohn et al. (2011, 2012) have been compared with actual O_3 data. In the EPA AQS database, hourly average O_3 concentrations are at times marked with various coding by the state or tribe entity responsible for collecting the data. One specific code is "RO". The code signifies that the governmental entity responsible for reporting the hourly O_3 data into the EPA's AQS database plans to submit a demonstration that the value(s) should be excluded from the NAAQS calculations for attainment purposes. The Fig. 3-33 below illustrates for a site in the Denver area (AQS ID 080590006) for May 2012 the relationship between the STT-S trajectories described above and calculated by Professor Heini Wernli (Institute for Atmospheric and Climate Science, ETH Zurich, Switzerland) and the "RO" codes embedded in the site's hourly data file in the AQS database. All hourly average O_3 concentrations in the AQS database from May 26 at 2000 through May 29 at 0100 local standard time (LST) were designated with an "RO" code. An hourly average O_3 concentration of 80 ppb was recorded on May 27, 2012 at 0300 LST, which would be 1000 GMT on the figure below. The STT-S "hits" appear to agree well with the "RO" designations in the AQS database for this site. Similar comparison agreements between STT-S trajectories and observed values have been described (Lefohn et al., 2011).

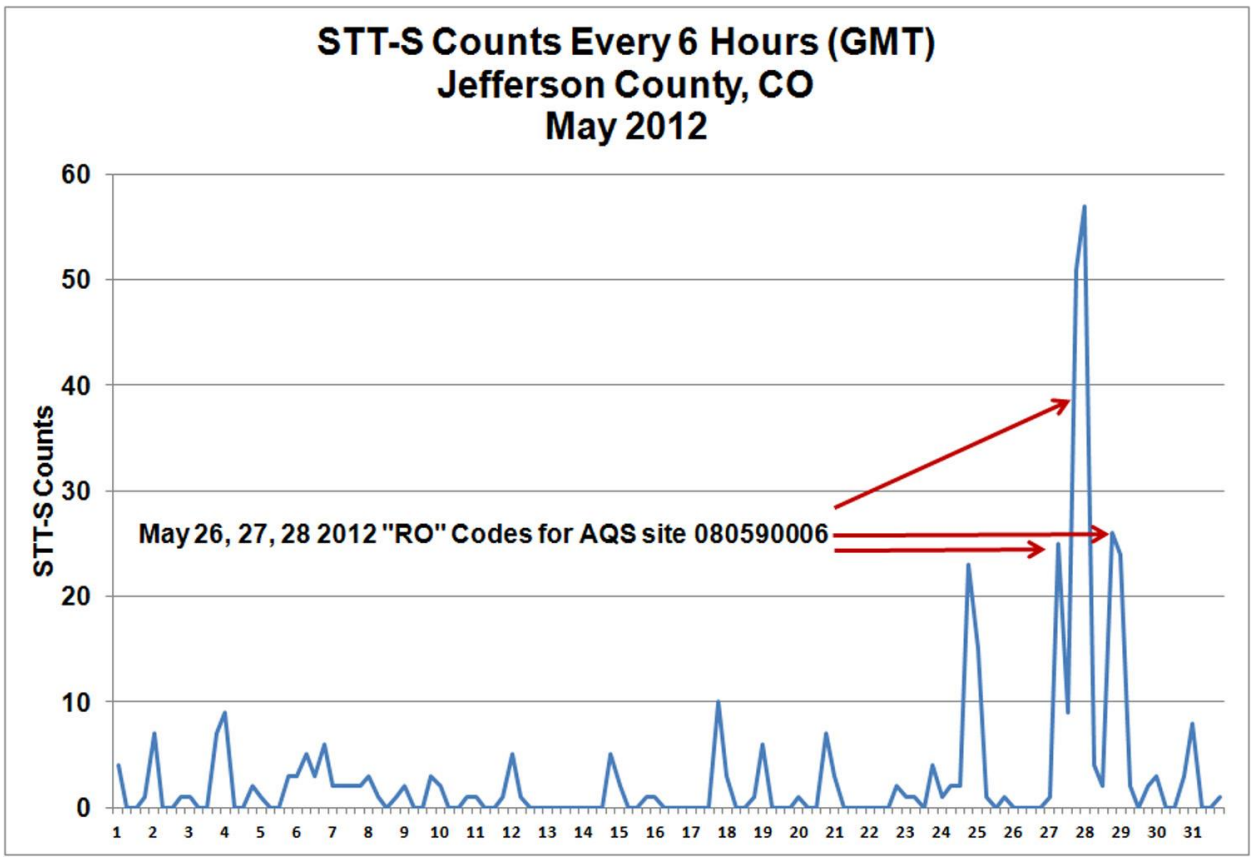


Figure 3-33. Relating STT-S counts for every six hours (GMT) with "RO" codes for a site in Jefferson County, Colorado (AQS ID 080590006) for May 2012. See Lefohn et al. (2011, 2012, 2014) for details how the STT-S values are estimated.

On page IS-14 of the draft EIS, the authors state that international transport from Asia has also been identified as a major source of precursors that contribute about 5 to 7 ppb to USB O_3 concentrations over the western U.S. (U.S. EPA, 2013b, 2006a, b). Ozone precursor emissions from China and other Asian countries have been estimated to have more than doubled in the period 1990−2010, and an estimated increase of 0.3 to 0.5 ppb/year of mid-tropospheric O_3 USB in spring over the western U.S. in the two decades after 1990 was largely attributed to a tripling of Asian NO_X emissions. However, after this period, trends in NO_X emissions from China, the largest O_3 precursor source in Asia, have declined as confirmed by rapidly decreasing satellite derived tropospheric NO_2 column measurements over China since 2012. Stringent air quality standards implemented in 2013 within China have markedly reduced national emissions.

On page IS-14 of the draft ISA, the authors note that other contributors (i.e., wildfires, lightning, global methane emissions, and natural biogenic and geogenic precursor emissions) to USB are either smaller or more uncertain than stratospheric and intercontinental contributions. EPA notes on page 1-1 of the draft ISA that an increasing trend of U.S. background concentration at high-elevation western U.S. sites before approximately 2010 now shows signs of slowing or even reversing, probably due to decreasing East Asian precursor emissions. I would urge caution in drawing conclusions about the anticipated effects in the U.S. as a result of emission reductions in China.

Lefohn et al. (2017) analyzed trends using hourly average O_3 concentrations from monitoring sites in Hong Kong and mainland China. The authors noted the following:

> The changes in O_3 concentration distribution at the Chinese sites were most commonly associated with shifts towards higher concentrations, with the result that the metrics either increased in magnitude or showed no trend. Although NOx emissions reductions occurred over a short time frame toward the end of the study period, mainland China and Hong Kong exhibited increasing trends in many of the exposure metrics. Although speculative, possible reasons for not observing significant trend reductions in the exposure metrics in China may be associated with the need for a longer period than six years (2010-2015) for emission changes to influence the metric trend patterns. In addition, the scarcity of monitoring stations could possibly contribute to lack of clear trend patterns. Year-to-year variability of meteorology could be a large factor in not observing decreases in the exposure metrics. In addition, further reductions in NOx levels may be required before decreasing trends are observed. At many of the Chinese sites, O_3 formation is sensitive to VOCs rather than NOx; VOCs have been increasing in mainland China (Ohara et al., 2007).

Lefohn et al. (2017) further noted

> In contrast to decreasing emissions in the EU and US, emissions of NOx have increased until recently in mainland China. Conversely, in Hong Kong, there have been large reductions in local emissions of both NOx and VOC since 1997. However, peak ambient O_3 concentrations have not decreased due to the contribution of long-range transport from increasing O_3 levels from mainland China (Xue et al., 2014). NOx emissions in China peaked around 2010-2011 and have since decreased (Duncan et al., 2016). Distribution changes at sites in China were most commonly associated with shifts towards higher concentrations.

There is inconsistency on the influence of long-range transport from Asia on western high-elevation O_3 monitoring sites. Long-range transport from Asia has not influenced trend patterns at all western US high-elevation O_3 monitoring sites. Not all high-elevation western U.S. sites have exhibited statistically significant trends during the springtime, when transport is expected to be highest from Asia. An evaluation of trend patterns of high-elevation western U.S. sites during the springtime, using the 4^{th} highest daily maximum 8-h concentration exposure metric, shows that some sites do not experience increasing trends over the period 2000-2014.

The draft ISA attributes increasing trend patterns observed at high-elevation western U.S. sites to long-range transport from Asia. As indicated above, I suggest that the authors indicate caution in drawing the strong conclusions currently in the document about the impact of long-range transport on western high-elevation O_3 monitoring sites, as well as anticipated changes as a result of emission reductions in Asia.

3.2.7 What do we know about the seasonal pattern of stratospheric-to-tropospheric transport to the surface (STT-S) and why is it important?

The EPA's White Paper (EPA, 2015) stated the following:

> Away from the earth's surface, O_3 can have an atmospheric lifetime on the order of weeks. As a result, background O_3, and to a lesser extent background O_3 precursors, can be transported long distances in the upper troposphere and be available to mix down to the surface when conditions are favorable. One of the largest natural sources of O_3 originates from production of O_3 in the stratosphere through interactions between ultraviolet light and molecular oxygen. O_3 exists in large quantities in the stratosphere and natural atmospheric exchange processes can transport stratospheric air into the troposphere. During certain meteorological conditions, discrete plumes of stratospheric air can be displaced far into the troposphere and impact ground-level O_3 concentrations. These events are called stratospheric intrusions and can result in relatively high USB levels of O_3 at the surface, especially at higher-elevation sites. Other natural sources of O_3 precursor emissions include wildfires, lightning, and vegetation. Biogenic emissions of methane, which can be chemically converted to O_3 over relatively long time scales, can also contribute to USB O_3 levels. Finally, manmade precursor emissions from other countries can contribute to the global burden of O_3 in the troposphere and to increased USB O_3 levels.

In addition, page 1-24, the draft ISA states:

> Deep stratospheric intrusions are common in the western U.S., impacting high elevation locations during the springtime. The incidence of tropopause folds is greatest in the early part (late winter and spring) of the year when synoptic-scale midlatitude cyclones are most active, occurring near upper level frontal zones where Rossby wave breaking is prevalent (Langford et al., 2017; Škerlak et al., 2015; U.S. EPA, 2013; Lin et al., 2012a).

Figs. 3-34, 3-35, 3-36, 3-37, and 3-38 illustrate as examples the daily maximum 8-h average concentration (MDA8) USB_{AB} estimates, observed daily MDA8 values, and the daily STT-S counts (i.e., hits) for Yellowstone National Park (WY), Jefferson County (CO), Rocky Mountain National Park (CO), Lassen Volcanic National Park (CA), and Sacramento (CA). For the Yellowstone National Park site, it appears that STT-S plays an important role during the April-October period. During the summer, when the STT-S "hits" are strongly reduced at the other three sites, USB_{AB} is slightly reduced at the high-elevation Jefferson and Rocky Mountain National Park sites and therefore, periods occur with "gaps" between observed values and USB_{AB}, likely attributable to anthropogenic sources. The amplitude of the "gap" varies strongly between the four sites. During rare events of STT-S > 0 in summer (e.g., Lassen, end of July and end of August), the two curves approach one another, indicating that STT-S episodes can also occur in summer with the result there is a close agreement between observed values and USB_{AB}. For the Sacramento site, STT-S events occur during the spring and fall months. Gaps (i.e., the difference between the observed and USB_{AB} concentrations) occur from mid-May through

September, indicating the apparent influence of anthropogenic sources. STT-S events occur across the US at all elevations with the result that USB_{AB} contributes in varying amounts (depending upon season and elevation of the site) to the observed O_3 concentrations across the US. USB_{AB}, while important in the high-elevation sites in the western US, is also important at low-elevation sites across the US (Lefohn et al., 2011, 2012, 2014).

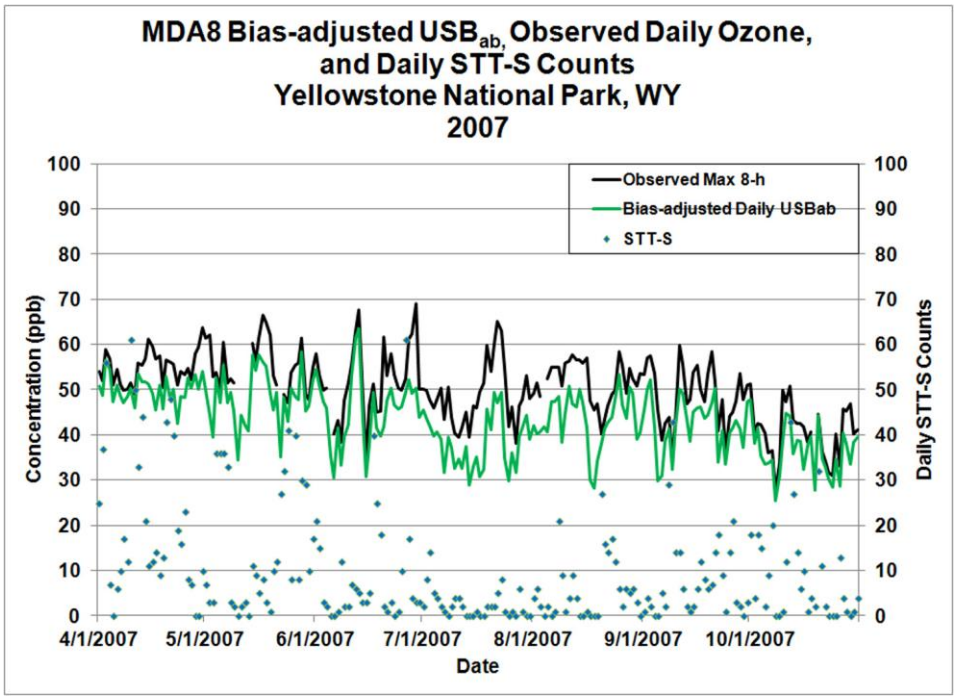


Figure 3-34. A comparison of the observed 8-h daily maximum concentration with the estimated bias adjusted US Background (USB_{AB}) 8-h daily maximum concentrations for Yellowstone National Park (AQS ID 560391011) for April-October 2007. The daily stratospheric-tropospheric transport to surface (STT-S) trajectories, as estimated by Professor Heini Wernli, Institute for Atmospheric and Climate Science, ETH Zurich, Switzerland, are overlaid with the daily O_3 values. Daily USB_{AB} 2007 values provided by the EPA. See Lefohn et al. (2011, 2012, 2014) for details how the STT-S values are estimated.

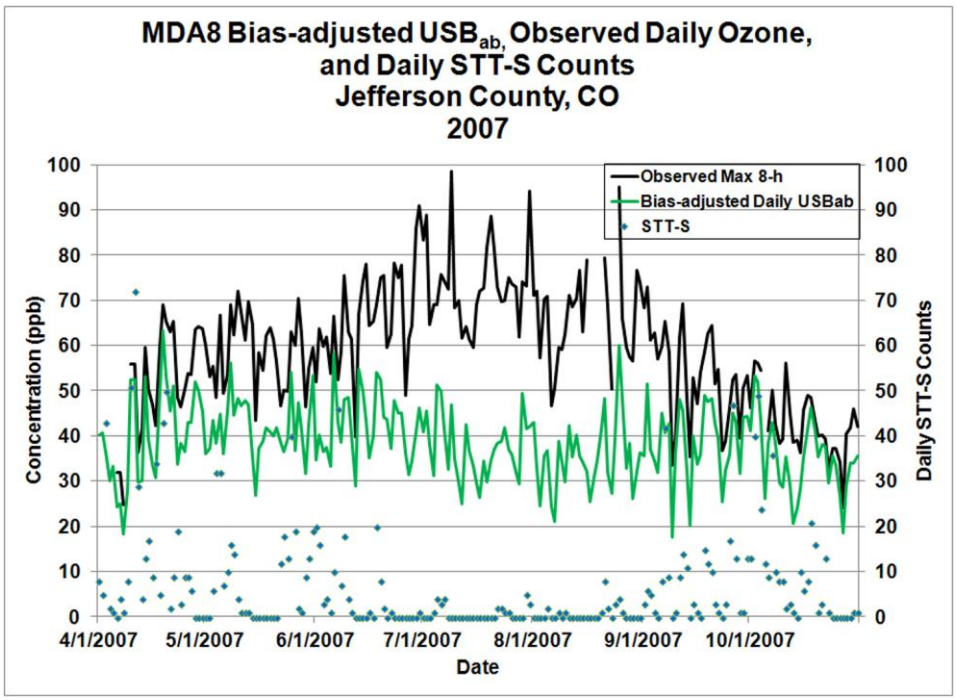


Figure 3-35. A comparison of the observed 8-h daily maximum concentration with the estimated bias adjusted US Background (USB_{AB}) 8-h daily maximum concentrations for Jefferson County, Colorado (AQS ID 080590006) for April-October 2007. The daily stratospheric-tropospheric transport to surface (STT-S) trajectories, as estimated by Professor Heini Wernli, Institute for Atmospheric and Climate Science, ETH Zurich, Switzerland, are overlaid with the daily O_3 values. Daily USB_{AB} 2007 values provided by the EPA. See Lefohn et al. (2011, 2012, 2014) for details how the STT-S values are estimated.

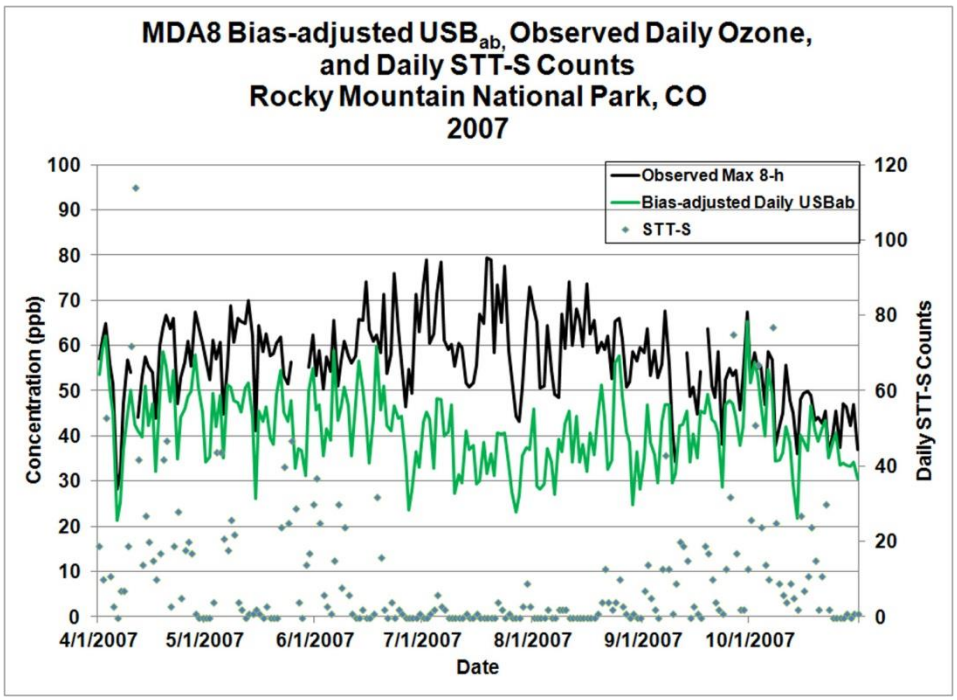


Figure 3-36. A comparison of the observed 8-h daily maximum concentration with the estimated bias adjusted US Background (USB_{AB}) 8-h daily maximum concentrations for Rocky Mountain National Park (AQS ID 080690007) for April-October 2007. The daily stratospheric-tropospheric transport to surface (STT-S) trajectories, as estimated by Professor Heini Wernli, Institute for Atmospheric and Climate Science, ETH Zurich, Switzerland, are overlaid with the daily O_3 values. Daily USB_{AB} 2007 values provided by the EPA. See Lefohn et al. (2011, 2012, 2014) for details how the STT-S values are estimated.

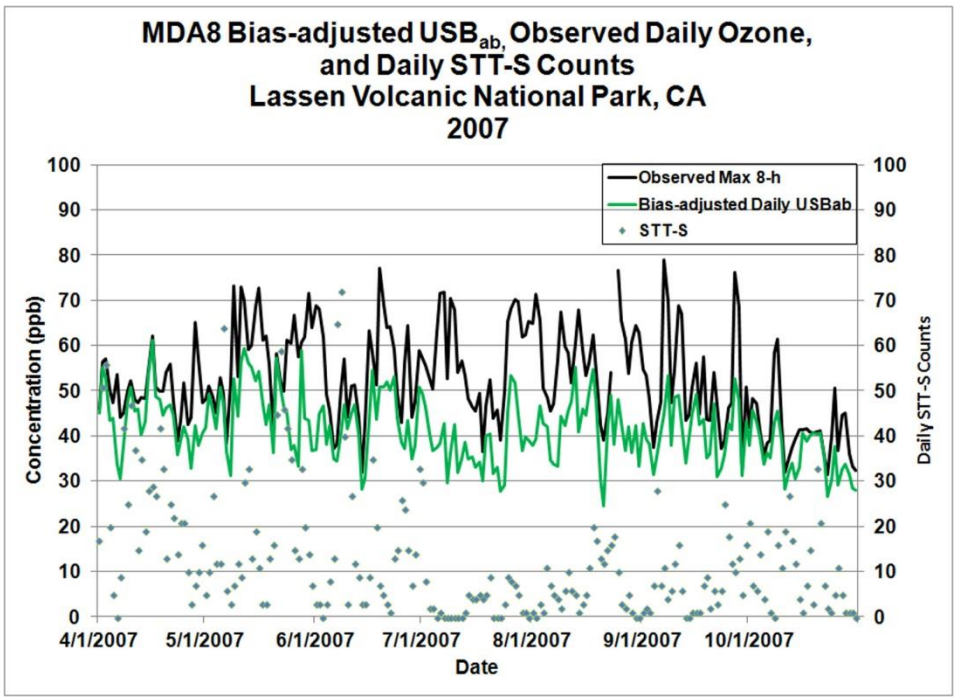


Figure 3-37. A comparison of the observed 8-h daily maximum concentration with the estimated bias adjusted US Background (USB_{AB}) 8-h daily maximum concentrations for Lassen Volcanic National Park (AQS ID 060893003) for April-October 2007. The daily stratospheric-tropospheric transport to surface (STT-S) trajectories, as estimated by Professor Heini Wernli, Institute for Atmospheric and Climate Science, ETH Zurich, Switzerland, are overlaid with the daily O_3 values. Daily USB_{AB} 2007 values provided by the EPA. See Lefohn et al. (2011, 2012, 2014) for details how the STT-S values are estimated.

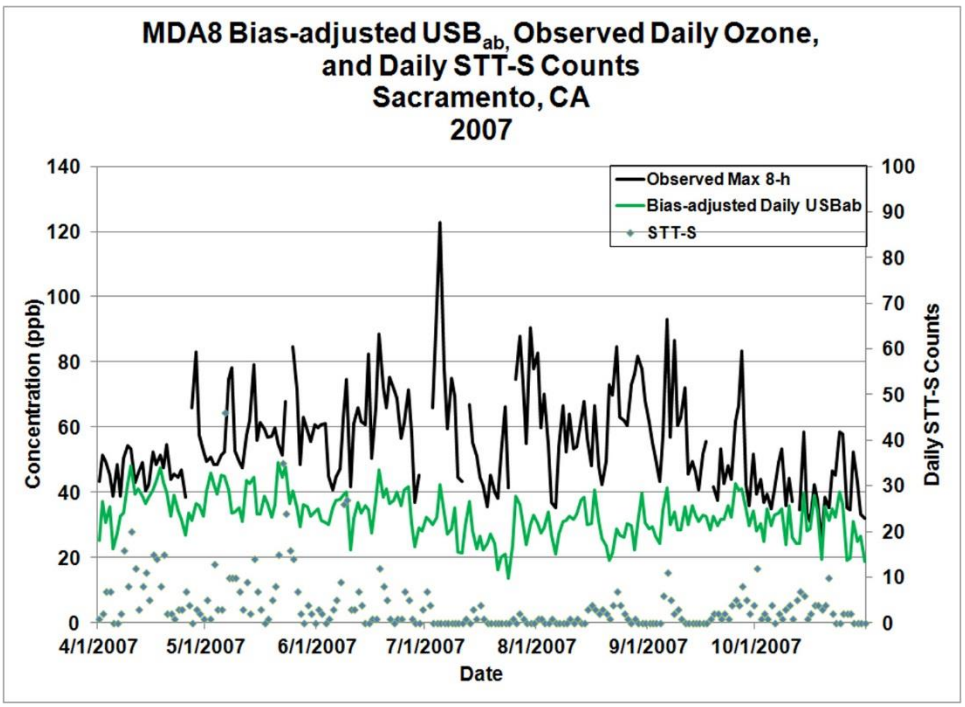


Figure 3-38. A comparison of the observed 8-h daily maximum concentration with the estimated bias adjusted US Background (USB_{AB}) 8-h daily maximum concentrations for Sacramento (AQS ID 060670012) for April-October 2007. The daily stratospheric-tropospheric transport to surface (STT-S) trajectories, as estimated by Professor Heini Wernli, Institute for Atmospheric and Climate Science, ETH Zurich, Switzerland, are overlaid with the daily O_3 values. Daily USB_{AB} 2007 values provided by the EPA. See Lefohn et al. (2011, 2012, 2014) for details how the STT-S values are estimated.

3.2.8 Future Research on Health

Areas of future health research are discussed in the draft PA (Section 3.6) starting on page 3-88. While normally I would reserve comment on Section 3.6 in the draft PA until I have prepared a separate set of comments that focuses on the draft PA, I believe it is important to integrate many of the areas covered in my comments on the draft ISA with the future health research needs so that the reader is able to access the materials presented earlier in my comments.

On page 3-88 of the draft PA, the authors note that a critical aspect of the Agency's consideration of the evidence and the quantitative risk/exposure estimates is the understanding of O_3 effects below the lowest concentrations studied in controlled human exposure studies, for longer exposures and for different population groups, particularly including people with asthma. The authors highlight areas for future health-related research, model development, and data collection activities to address these uncertainties and limitations in the current scientific evidence. The items identified are as follows:

1. An important aspect of risk assessment and characterization to inform decisions regarding the primary standard is our understanding of the exposure-response relationship for O_3-related health effects in at-risk populations. Additional research is needed to more comprehensively assess risk of respiratory effects in at-risk individuals exposed to O_3 in the range of 40 to 80 ppb***, and lower*** (emphasis added), for 6.6 hours while engaged in moderate exertion.

As noted in the draft PA, the lower hourly average O_3 concentrations shift from the lower values toward the mid-range as emissions are reduced. The upward shifting of the hourly average concentrations from the low-end of the distribution is indicative of background O_3 becoming more and more important in the 25-55 ppb range. Thus, suggesting that researchers utilize hourly average O_3 concentrations that are in the background part of the distribution may not provide helpful information for the standard-setting process. I would suggest that additional focus be on the 50-70 ppb range.

2. Epidemiologic research continues to be important to consideration of the public health impacts of O_3. However, there remains a need to continue to examine and improve analytical approaches, and to better understand the role of co-pollutants, as well as temperature, in contributing to potential confounding or effect modification in epidemiologic models.

The heterogeneity that is evident in the epidemiologic research results needs to be better explained. Temperature and the role of co-pollutants may help explain the heterogeneity observations.

3. Epidemiologic studies assessing the influence of "long" or "short" O_3 exposures is complicated by the exposure history of study populations. Further, existing studies generally focus on either long-term or short-term exposure separately, thereby making it difficult to assess ***whether a single exposure versus a longer exposure, or a combination of both single and longer exposure, influence health outcomes of the study subjects*** (emphasis added). Studies that include a longer assessment period and can simultaneously assess the impact of these various exposure durations are needed.

While it may appear that to reduce acute O_3 health effects requires a short-term standard, such as the 4th highest 8-h daily maximum exposure metric and that a long-term average concentration is required to reduce chronic O_3 health effects, such is not necessarily the case. In 2015, the EPA believed that by implementing a control strategy that reduces the higher concentrations that concentrations of concern for "chronic" effects would also be reduced (Federal Register, 2015 – page 65399). The US EPA (Federal Register, 2015 – page 65358) commented on how the Agency chose to reduce "chronic" and "acute" O_3 exposures for the protection of human health. The Agency believed that the reduction of the repeated occurrences of exposures of concern would reduce both "chronic" and "acute" health effects. The EPA stated:

> ...This point was also highlighted by some commenters who advocated for a level of 60 ppb, based on the discussion of O_3-induced inflammation in the proposal. In particular, this latter group of commenters highlighted discussion from the

> proposal indicating that "[i]nflammation induced by a single O_3 exposure can resolve entirely but, as noted in the ISA (U.S. EPA, 2013a, p. 6-76), 'continued acute inflammation can evolve into a chronic inflammatory state'" (e.g., ALA et al., p. 48). Consistent with these comments, and with her consideration of estimated exposures of concern in the proposal, the Administrator judges that the types of respiratory effects that can occur following exposures of concern, particularly if experienced repeatedly, provide a plausible mode of action by which O_3 may cause other more serious effects. ***Because of this, as in the proposal, the Administrator is most concerned about protecting against repeated occurrences of exposures of concern*** (emphasis added).

The EPA then commented (Federal Register, 2015 – page 65358) on the reduction of the higher concentrations and how these reductions not just affect the highest MDA8 concentrations, but also those values that are below these highest levels. In other words, by reducing the peak exposures, there is a cascading of the upper end of the distribution of O_3 concentrations down toward the mid-level values. The EPA stated as follows:

> ...In addition, though the available information does not support the identification of specific benchmarks below 60 ppb that could be appropriate for consideration for at-risk populations, and though CASAC did not recommend consideration of any such benchmarks, the EPA expects that a revised standard with a level of 70 ppb will also reduce the occurrence of exposures to O_3 concentrations at least somewhat below 60 ppb (U.S. EPA, 2014a, Figures 4-9 and 4-10). Thus, even if some members of at-risk populations may experience effects following exposures to O_3 concentrations somewhat below 60 ppb, a revised level of 70 ppb would be expected to reduce the occurrence of such exposures. Therefore, the EPA has considered O_3 exposures that could be relevant for at-risk populations such as children and people with asthma, and does not agree that controlled human exposure studies reporting respiratory effects in healthy adults following exposures to 60 ppb O_3 necessitate a standard level below 70 ppb.

Thus, given EPA's conclusions reached in the 2015 O_3 rulemaking process as summarized above, it is suggested that both epidemiological researchers, as well as policy makers, understand that a long-term epidemiological study does not have to use metrics that are based on long-term averages. Rather, long-term epidemiological studies should also consider that the long-term effects may be associated with repeated acute exposures. Lefohn et al. (2010) described a sigmoidal weighting scheme for hourly average O_3 concentrations. The weighting scheme addresses the nonlinearity of response (i.e., the greater effect of higher concentrations over the mid- and low-range values) on an hourly basis. The weighting might be relevant for those who wish to develop daily O_3 exposure metrics that could be used to integrate cumulative exposures over time. The W90 weighting scheme, different in form than the W126 metric currently used for assessing O_3 risk to vegetation, is shown in Fig. 3-39 below. The form of the W90 index is $\Sigma\ w_i \times C_i$ with weight $w_i = 1/[1 + M \times exp\ (-A \times C_i/1000)]$, where $M = 1400$, $A = 90$, and where C_i is the hourly average ozone mixing ratio in units of ppb. The W90 index has units of ppb-hrs.

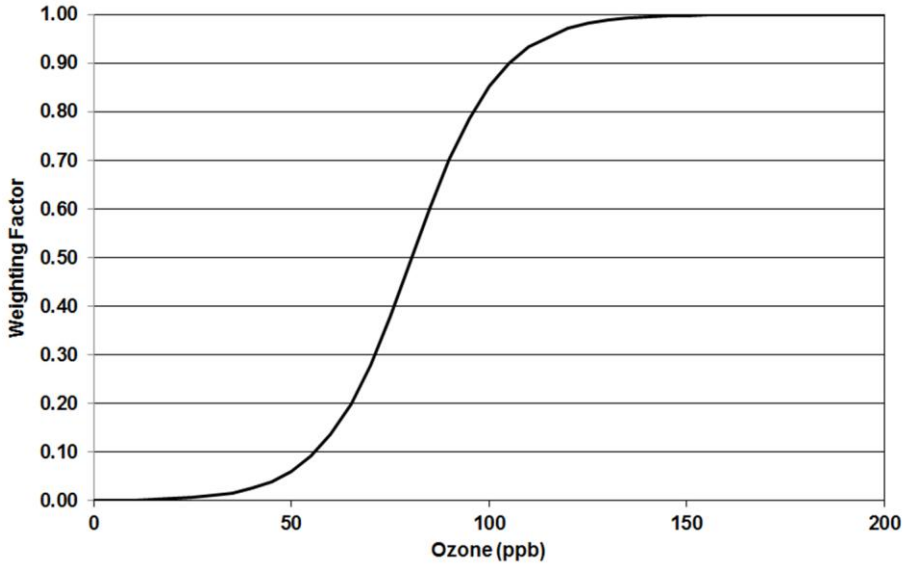


Figure 3-39. The weighting applied to hourly average ozone values for the calculation of the W90 exposure index (see Lefohn et al., 2010).

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