Comments on Draft Review of the Ozone National Ambient Air Quality Standards

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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 presented in the Draft Review of the Ozone National Ambient Air Quality Standards. His comments are submitted into the Ozone Docket (No. EPA-HQ-OAR-2018-0279) for the purpose of providing scientific clarification 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 that are important for the O_3 standard-setting science assessments. 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 in its 2015 O_3 rulemaking for the 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 substitute for the W126 index to protect vegetation; the EPA continues to utilize 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. His research results are cited in the current EPA Integrated Science Assessment and the Policy Assessment documents. 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 was a member of the first Steering Committee (2014-2019) 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. On September 25, 2019 and October 31, 2019, the only drafts of the EPA's Integrated Science Assessment for Ozone (ISA) (EPA, 2019a) and Policy Assessment (PA) (EPA, 2019b) documents were issued, respectively. The compressed schedule during this rulemaking cycle resulted in the simultaneous preparation of both documents. In contrast, during the 2015 ozone (O_3) rulemaking activities, additional time was permitted for the review and accurate integration of the various rulemaking documents. During that rulemaking cycle, 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. In his April 1, 2020 letter to Dr. Louis Anthony Cox, Jr., Chair of CASAC, the Administrator (EPA, 2020c) noted that the CASAC had raised several important issues with the draft Ozone ISA. As noted by the Administrator, the CASAC found that the draft Ozone ISA "does not provide a comprehensive, systematic assessment of the available science relevant to understanding the public health impacts of changes in ambient concentrations of ozone." The Administrator acknowledged that some of the CASAC comments and adjustments would be addressed in the final version of the ISA, while other CASAC comments would require additional time to complete. Because of time limitations noted by the Administrator in his April 1, 2020 letter to CASAC, in some cases, the final version of the ISA did not necessarily reflect the latest state-of-science evidence. Because the ISA and PA are tightly linked together, the inadequacies in the ISA (EPA, 2020a) result in some of the PA (EPA, 2020b) findings not necessarily reflecting the best science available (e.g., critically evaluating information important for assessing margin of safety considerations, adequately estimating background O_3 levels, clearly stating emission reduction strategies for reducing chronic and acute human health effects, and recognizing the limitations of using the current form and level of the primary standard to protect vegetation). Several of these inadequacies are addressed in the written comments contained within this document.

2. Two key fundamental principles 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**. For human health, this principle is based on an important series of human health clinical studies published by Hazucha et al. (1992) and Adams (2003, 2006a, b). These controlled human health clinical studies showed that greater O_3 peak responses were observed in stepwise and triangular (smooth increases and decreases in concentration) exposures rather than in constant concentration exposure protocols. (Sections 1.3 and 2).

3. For vegetation, the first fundamental principle is supported by key research results reported in the 1980s and 1990's. In the most current ISA (EPA, 2020a), the Agency

continues to conclude, based on experimental studies, 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, where substantial reductions over the years in the higher hourly average O_3 concentrations in the Los Angeles area occurred, provide independent confirmation of the experimental studies for the greater importance of the higher hourly average O_3 concentrations in influencing vegetation effects. (Sections 1.3 and 2.2).

4. One result of the first fundamental 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. (Sections 1.3 and 2.1)

5. In implementing its Air Quality Index (AQI) reported across the U.S., EPA recognizes the importance of the higher O_3 concentration. 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. (Sections 1.3 and 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-h concentrations.

6. The Administrator notes (Federal Register, 2020 – page 49842) that in the review completed in 2015 that an 8-h averaging time remained appropriate for addressing health effects associated with short-term exposures to ambient air O_3 and that it could effectively limit health effects attributable to both short- and long-term O_3 exposures (80 FR 65348, October 26, 2015). Simply stated, by reducing *the higher part of the distribution of hourly average concentrations* (not just the peak hourly values), the EPA Administrator believed in 2015 that the risk to human health and vegetation would be reduced by reducing the hourly average O_3 concentrations at the upper part of the

distribution curve. In addition, the EPA in its 2015 decision (Federal Register, 2015 – pages 65358 – 65359) anticipated that a revised standard with a level of 70 ppb would also reduce the occurrence of exposures to O_3 concentrations at least somewhat below 60 ppb based on its modeling results in the 2014 Health Risk and Exposure Assessment document (EPA, 2014b, Figs. 4-9 and 4-10). Thus, even if some members of at-risk populations might experience effects following exposures to O_3 concentrations somewhat below 60 ppb, the Administrator believed in 2015 that a revised level of 70 ppb would be anticipated to reduce the occurrence of such exposures. Thus, the EPA believed it had considered O_3 exposures that could be relevant for at-risk populations, such as children and people with asthma, and did not agree in 2015 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. 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 revised O_3 standards. (Sections 1.3, 1.7, and 3.1).

7. The second key 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**. As O_3 precursor emissions are reduced at specific locations across the U.S. to attain the current O_3 NAAQS, not all hourly average O_3 concentrations at these locations shift downward. In fact, the lower hourly average O_3 concentrations shift upward as described in the next two items. The second fundamental principle is supported using empirical data, as well as atmospheric chemistry/meteorological modeling results published in the peer-reviewed literature. (Sections 1.3, 1.8, and 3).

8. As emissions are reduced to meet the O_3 NAAQS, a compression of the higher and lower hourly average O_3 concentrations occurs at many sites in the U.S. The higher individual 8-h daily maximum (MDA8) values are reduced downward toward the mid-level concentrations, while the lowest MDA8 values ***increase*** toward the mid-level values. As an example, for an urban-influenced site in 1985 in Jefferson County, Kentucky, there were frequent occurrences of high and low hourly average O_3 concentrations (Fig. ES-2). The site in 1985 appeared to be influenced by NO titration of O_3 by NO_x emissions because of the occurrence of frequent low hourly average concentrations. The distribution of hourly average data for the same Kentucky site in 2017 is shown in Fig. ES-3. Although the site is still influenced by anthropogenic sources, the highest hourly average O_3 concentration has been reduced from 112 ppb (experienced in 1985) to 77 ppb (experienced in 2017). In addition, a shift of the lower concentrations toward the mid-level values has occurred. The shift is associated with less titration of O_3 by NO as reduction in NO_x emissions occur (Lefohn et al., 1998; EPA, 2014b; Simon, 2015; Lefohn et al., 2017, 2018). The reduction of O_3 precursors results in both the high and the low concentrations shifting toward the mid-level values, resulting in a compression of the distribution of hourly average concentrations. (Sections 1.3 and 3.1).

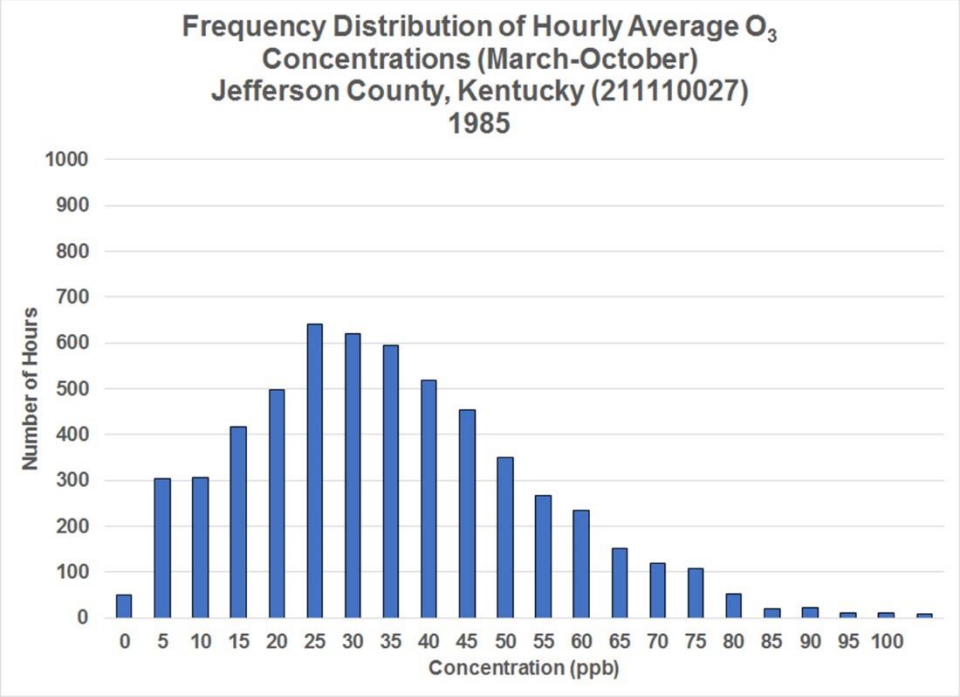


Figure ES-2. Frequency distribution of the hourly average O_3 concentrations in 1985 for an urban-influenced site in Jefferson County (KY) (AQS ID 211110027) monitoring station. Source of data is from the EPA's AQS database.

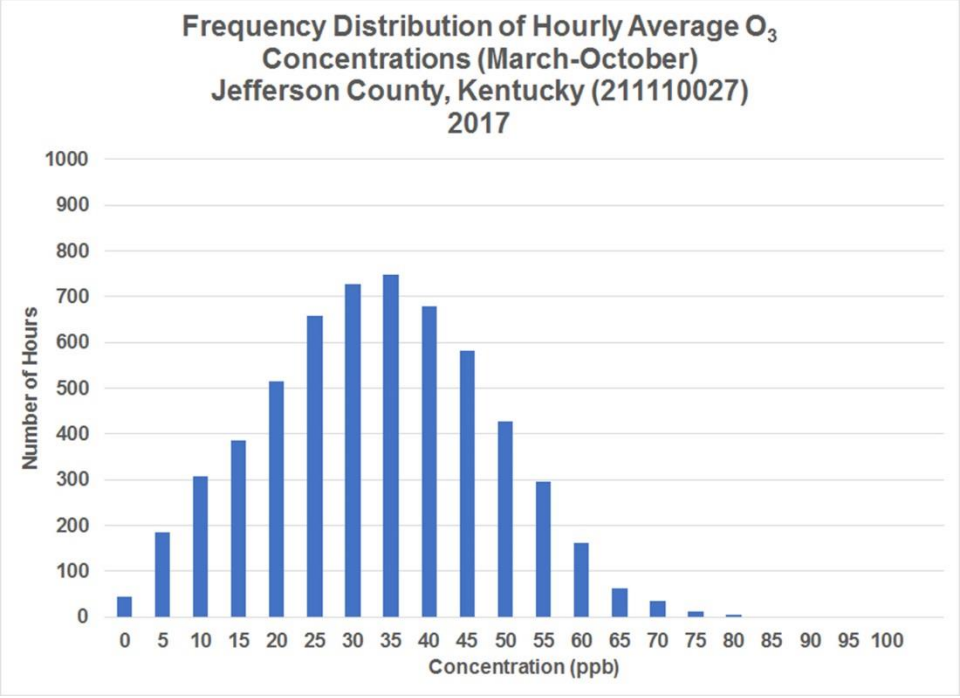


Figure ES-3. Frequency distribution of the hourly average O_3 concentrations in 2017 for an urban-influenced site in Jefferson County (KY) (AQS ID 211110027) monitoring station. Source of data is from the EPA's AQS database.

9. Because of the unfortunate COVID-19 pandemic, many countries around the world during the spring (northern hemisphere) and fall (southern hemisphere) of 2020 instituted immediate lockdown orders. As a result of these orders, anthropogenic emissions were severely reduced. Recognizing the opportunity to investigate how severe emission reductions influenced air pollution concentrations, researchers documented the air quality changes associated with this "natural experiment." One important result of the 2020 lockdown was documentation about the changes that occurred in the distribution of hourly average O_3 concentrations as emissions were reduced. During the 2020 COVID-19 lockdown, scientists characterized these changes by applying different O_3 exposure metrics (e.g., 24-h daily averages, median daily maximum 8-h concentrations, etc.). Some of the exposure metrics focused on the lower part of the distribution of hourly average concentrations (e.g., 24-h average concentrations), while other metrics focused on the upper part of the distribution (daily maximum 8-h average concentrations). Those investigators who used metrics focused on the higher concentrations reported *decreasing* O_3 concentrations during lockdown; those who applied metrics focused on the lower end of the distribution reported *increasing* O_3 concentrations. The lockdown that occurred during the spring in the U.S. resulted in large emission reductions of O_3 precursors. The results described by Sommer et al. (2020) for U.S. sites in a National Public Radio analysis, as well as supplemented by the analysis of 52 U.S. O_3 monitoring sites described in these comments, illustrate the decrease in daily maximum 8-h average concentrations at many locations. One investigator, using exposure metrics not focused on the higher hourly average concentrations during the lockdown period, reported mixed patterns for 28 O_3 monitoring sites in the U.S. As noted, some of the researchers who characterized O_3 monitoring sites around the world during lockdown applied metrics focused on the lower end of the distribution. Some of these investigators reported that increasing O_3 concentrations appeared to be related to the reduction of NO titration of O_3. The result of this "natural experiment" provides additional evidence that emission reductions results in the less biologically important hourly average concentrations increasing. This observation supports the second fundamental principle 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**. (Section 1.8).

10. The compression of the distribution of hourly average O_3 concentrations results in annual average or median concentration values ***increasing*** at some sites. For example, in Fig. ES-4 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 8-h O_3 exposures lower than the two urban sites. When the annual average is plotted (Fig. ES-5) for the same period with the same data, the ordering of the sites from the highest to the lowest annual average concentrations shows a different pattern. 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 annual average

exposure. Clearly, the ordering of the sites from highest to lowest exposures observed when using the 8-h metric is much different than the ordering when the annual average index is used. The increase in the annual average concentration values, even though emissions are being reduced, is associated with the low end of the distribution increasing due to less titration of O_3 by NO. 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 U.S. 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 annual 4th highest daily maximum 8-h average concentration metric. (Sections 1.3 and 3.1).

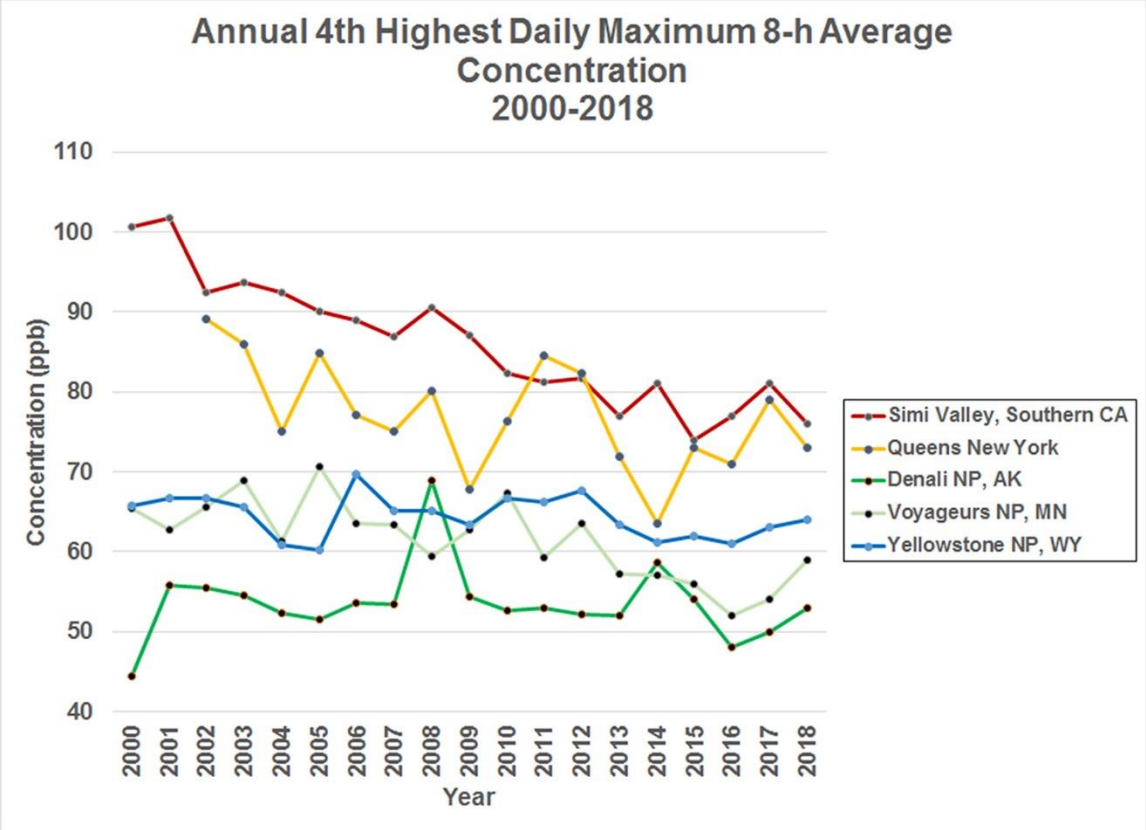


Figure ES-4. The annual 4th highest daily maximum 8-h average O_3 concentration for the period 2000-2018 for Simi Valley, CA (061112002), Queens New York, NY (360810124, Denali National Park, AK (020680003), Voyageurs National Park, MN (271370034), and Yellowstone National Park, WY (560391011). Source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

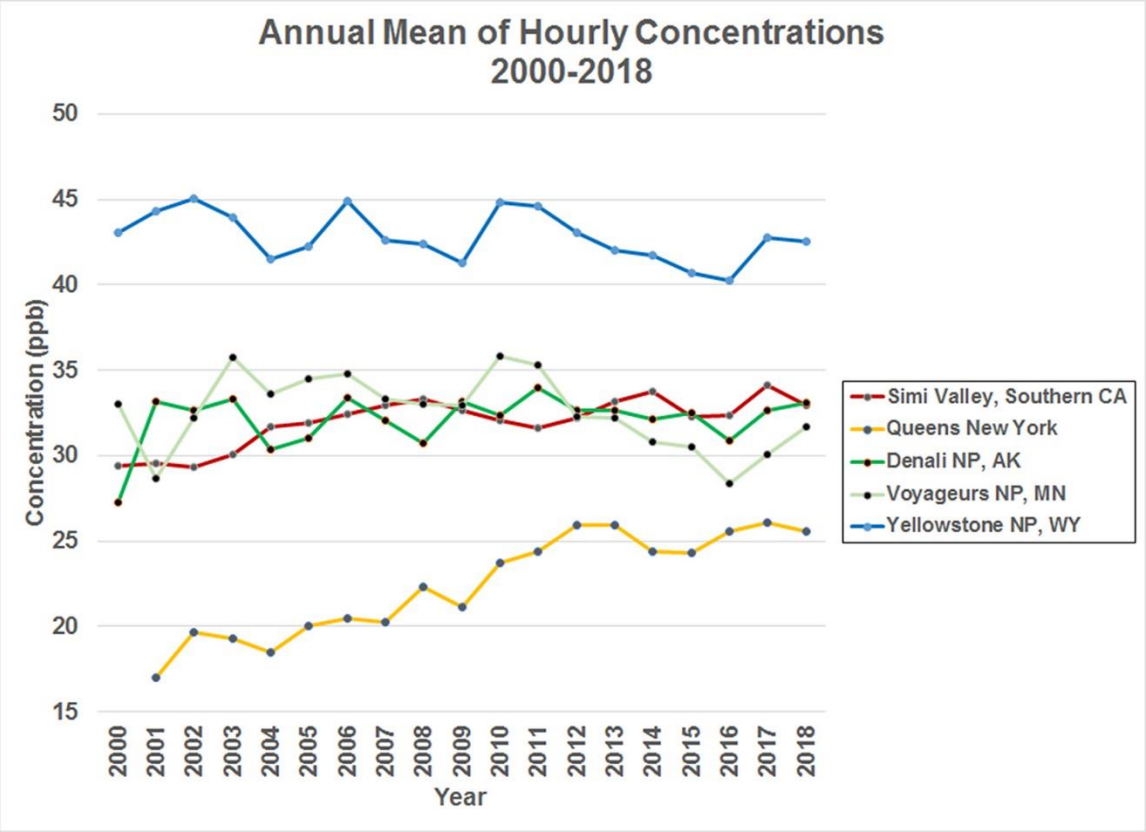


Figure ES-5. The annual mean of the hourly average O_3 concentrations for the period 2000-2018 for Simi Valley, CA (061112002), Queens New York, NY (360810124, Denali National Park, AK (020680003), Voyageurs National Park, MN (271370034), and Yellowstone National Park, WY (560391011). Source of data is from the EPA's AQS database.

11. One does not have to select necessarily an annual average or seasonal average concentration metric to estimate "chronic" (i.e., long-term) human health effects from O_3 exposures. Based on empirical data, annual average concentrations increase as emissions are reduced. The biologically important higher hourly average O_3 concentrations are reduced as emissions are reduced. In its review of the 2015 O_3 NAAQS rulemaking, the Administrator noted that an 8-hour averaging time remained appropriate for addressing health effects associated with short-term exposures to ambient air O_3 and that it could effectively limit health effects attributable to both short- and long-term O_3 exposures. (Sections 1.4 and 2.4).

12. The use of the W126 metric as a secondary standard to protect vegetation has received strong support from CASAC in previous reviews (Henderson, 2006; Samet, 2010; Frey, 2014). Samet (2010), in summarizing CASAC's comments on the reconsideration of the O_3 NAAQS by the Obama Administration, noted that in recommending the W126 for the secondary welfare standard, the Agency acknowledged the distinction between the effects of acute exposures to O_3 on human health and the effects of chronic O_3 exposures on

welfare, namely that vegetation effects were more dependent on the cumulative exposure to, and uptake of, O_3 over the course of the entire growing season. At the time, CASAC pointed out that the Agency was responding to the clear need for a secondary standard that was different from the primary standard in averaging time, level, and form. In 2010, the EPA proposed to set the level of a proposed W126 secondary standard within the range of 7-15 ppm-hours. In 2011, President Obama requested that the EPA withdraw its reconsideration of the O_3 standards, which included the proposed W126 secondary O_3 standard. In its 2015 O_3 NAAQS decision, the Administrator chose to use the current form of the human health standard and not the W126 index as the form and averaging time for the secondary standard. In August 2019, the U.S. Court of Appeals for the District of Columbia Circuit (DC Circuit) addressed arguments regarding the adequacy of the EPA Administrator's 2015 decision to use the current form of the O_3 human health standard instead of the W126 metric as the secondary O_3 NAAQS. One aspect of the Court's August 2019 decision was to question EPA's decision to use a 3-year average of the W126 index rather than a 1-year annual W126 metric to protect vegetation. The Court ruled that it lacked any basis to assess the reasonableness of EPA's actions to use the current form of the O_3 NAAQS instead of the W126 because EPA never explained why it was reasonable to focus on a 3-year average of the W126 index instead of an annual W126 value. In the Draft Review of the Ozone National Ambient Air Quality Standards document (Federal Register, 2020), the Agency attempted to respond to the Court on the justification for the use of a 3-year average of the W126 instead of an annual W126 by including examples of analyses from a multi-year study (King et al., 2005) described in the PA (EPA, 2020b). The EPA (Federal Register, 2020) cautioned that it might be premature to use the data from the King et al. (2015) study to draw a broad interpretation about whether the 3-year average of the W126 offers the same level of protection to vegetation as the use of an annual W126 index. One needs to be careful in drawing a broad interpretation of EPA's use of the King et al. (2005) data. If the Agency were to draw a broad interpretation from its analyses of the King et al. (2005) data, one might conceivably conclude, if the 3-year average of the W126 values were the same at two different sites, no difference in the predicted vegetation effects would be anticipated if an exposure regime at Site 1 over 3 years experienced many high and low hourly average concentrations, while a different exposure regime at Site 2 contained many mid-level hourly average concentrations and infrequent high and low hourly values. This assumption would contradict EPA's conclusion that the higher hourly average O_3 concentrations, which the W126 index weights more than the mid- and low values, are important when assessing adverse vegetation effects (EPA, 2020a, b). The frequency and magnitude of the higher hourly average concentrations play an important role for estimating vegetation effects over a growth season. It appears that the EPA has not sufficiently responded to the Court's concern about the selection of the 3-year average of the W126 metric instead of the annual W126. The use of the 3-year average of the W126 metric compromises the protection to vegetation offered by using an annual W126 index. (Section 1.9).

13. As noted in Item (12) above, in its 2015 O_3 NAAQS decision, the Administrator chose not to use the W126 index as the form and averaging time for the secondary welfare standard. The Agency found that O_3 exposure levels associated with the existing form

and averaging time were "highly correlated" to a 3-year average of the W126 index. In its August 2019 decision, the Court was concerned about the reasons that the EPA selected the 3-year average of the 4th highest daily maximum 8-h concentration instead of the W126 index. Although the EPA in the PA (EPA, 2020b) devotes a considerable amount of time illustrating the statistical relationship between the annual and 3-year average of the W126 metric with the current form of the standard, the 2014 CASAC (Frey, 2014) noted that "… the correlative similarity between the current standard and a level of the W126 index of 15 ppm-hrs must not be interpreted to mean that just meeting the current standard is equivalent to just meeting a W126 level of 15 ppm-hrs…" The 2014 CASAC (Frey, 2014) understood that correlative similarity did not mean that the current form of the standard could be substituted for the biologically relevant W126 index for the protection of vegetation. The exposure-response relationships developed for the agricultural crop and tree seedling estimates described in the ISA (EPA, 2020a) and PA (EPA, 2020b) are based on the experimental data reported in the literature. The W126 exposure-response relationships developed from the experimental data reflect the unique patterns of hourly average O_3 concentrations applied in the crop and tree growth experiments. Vegetation researchers who developed the exposure-response models realized that the use of an average concentration, such as the 4th highest daily maximum 8-h average concentration, reduced the ability to predict the cumulative effects associated with the patterns of the hourly average concentrations used in the experimental exposure regimes. The use of exposure indices that average hourly O_3 values (e.g., the annual 4th highest daily maximum 8-h average concentration) compromises the ability to apply those exposure-response relationships, which are based on experimental hourly exposures, to ambient concentrations recorded at O_3 monitors across the U.S. for assessing vegetation effects. The 3-year average of the 4th highest daily maximum 8-h average concentration metric is not a cumulative index and therefore, will not provide adequate information to protect against those regimes that elicit adverse vegetation effects. The same 3-year average of the 4th highest daily maximum 8-h average concentrations (e.g., 70 ppb) can represent different distributions of hourly average concentrations and therefore, different 1-year W126 values as illustrated in various figures presented in Appendix 4D in the PA (EPA, 2020b). This implies that while the correlative similarity between the current standard and a level of the W126 index may exist, the 3-year average of the 4th highest daily maximum 8-h concentration exposure index cannot adequately represent the distributions of hourly average O_3 concentrations responsible for vegetation injury and damage. The 2014 CASAC recognized the deficiencies in using the 8-h metric to protect vegetation and therefore, recommended the annual W126 index. The use of the 3-year average of the 4th highest daily maximum 8-h average concentration as a substitute for the 1-year W126 index is inadequate for protecting vegetation from those hourly average O_3 concentrations most important in eliciting adverse effects. The current 8-h form of the primary standard is used as a blunt tool for a job that requires a more precise object (i.e., the W126 index) for solving a major task: the protection of vegetation across the U.S. ***Based on items (12) and (13), the best protection for vegetation effects associated with O_3 exposures is to adopt the 1-year W126 exposure metric as the form of the secondary standard, which is different in averaging time, level, and form of the human health primary standard.*** (Section 1.9).

14. The Court of Appeals for the District of Columbia Circuit (DC Circuit) in its August 19, 2019 decision addressed arguments regarding considerations of background O_3 concentrations, and socioeconomic and energy impacts. Regarding background O_3, the Court rejected the argument that the EPA was required to take background O_3 concentrations into account when setting the NAAQS. The Court found that the text of the Clean Air Act 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 the EPA did not act unlawfully or arbitrarily or capriciously in setting the 2015 NAAQS without regard for background O_3. (Section 1.5).

15. While it is believed that background O_3 currently is not a consideration in the setting of the level of the O_3 standard, background O_3 plays an important role in influencing human health effects risk assessments. The human health risk and exposure assessments play an important role in the margin of safety determinations. Background O_3 concentrations in the low- and mid-level part of the distribution of concentrations make up a large fraction of the total ambient O_3 levels and potentially can influence those human health risk assessments associated with the margin of safety determinations for the setting of the primary O_3 NAAQS. (Section 1.6).

16. As emissions are reduced, besides the compression of the high and low concentrations toward the mid-level values, models predict that the highest concentrations, which normally have occurred in the past during the summer months, shift at some sites in the U.S. from the summer months to the March-June months. Besides modeling results, the data in the EPA's AQS database indicate sites where maximum concentrations shift from summer to spring months. There are also sites across the U.S. 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. (Sections 3.1.2 and 3.2.9).

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

18. In the ISA (EPA, 2020a, page 1-53), EPA states that background O_3 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 metric rather than an average exposure index, 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, at times 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 assessment. (Section 3).

19. The authors note that the term **US background (USB)** is used to assess background O_3. No clear reason is provided in either the ISA (EPA, 2020a) or the PA (EPA, 2020b) 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 PA. A key point made in the ISA (page 1-56) 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 O_3*** (Dolwick et al., 2015). The selection of USB rather than apportionment-based U.S. background (USB_{AB}), as well as not performing bias adjustments to the USB estimates, appear to be important concerns about the adequacy of the background O_3 modeling results presented in the PA. (Section 3).

20. The 2013 Ozone ISA (EPA, 2013) reported higher seasonal mean USB and NAB concentration estimates in spring than in summer for most regions of the U.S. EPA notes in the current ISA (EPA, 2020a, page 1-65) that while some new results are consistent with this pattern, other results suggest that summer USB O_3 concentrations can be comparable to or greater than spring concentrations. The ISA did not resolve the conflicting conclusions about when seasonal mean background O_3 is greatest. Data in the EPA's AQS database indicate sites where maximum concentrations have shifted from summer to spring months. There are also sites across the U.S. where the highest O_3 exposures occur in the spring independent of emission reductions. At some National Park Service (NPS) sites, the highest O_3 exposures across the U.S. occur during the springtime (March to mid-June). The EPA (2014c, page 7A-12) provided the highest 3-month W126 values and the timeframe corresponding to those W126 exposures for the Parks for the period 2006-2010. Several of the O_3 monitors in the Parks experienced their highest W126 exposures during the spring months (defined as March, April, May or April, May, June) period. In the 2015 NAAQS rulemaking (Federal Register, 2015 – page 65416), the EPA determined that the lengthening of the O_3 monitoring seasons in 32 states and the District of Columbia was appropriate. The Agency indicated that ambient O_3 concentrations in these areas could approach or exceed the level of the NAAQS, more frequently and during more months of the year compared with the length of the O_3 seasons prior to 2015. The EPA described the results of its analysis (Rice, 2014) and extended the seasons for specific states and the District of Columbia. In Section 3.2.8, additional material is provided that identifies the March-June occurrences of the highest O_3 exposures. The pattern when the highest exposures occur is important for validating model performance estimating background O_3 concentrations. There continues to be strong evidence, as supported in the literature, that background O_3 across the U.S. 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. (Sections 3.2.4 and 3.2.8),

21. The USB modeling results described in the PA (EPA, 2020b) indicate the following seasonal patterns: The natural contribution has a single maximum in late summer in the West, whereas, in the East there is evidence of two peaks— the largest in late Spring and a second peak in early Fall (page 2-48). The current analysis as described in the PA (EPA, 2020b, page 2-64) indicates that natural and U.S. anthropogenic O_3 contributions peak during the traditional O_3 season (May through September), while long-range intercontinental transport of international O_3 (i.e., contributions from China, India etc.) peaks in the spring (February through May). Reviewing Fig. ES-6, the total of the natural and international components show for the West that March-August appears to be the period of highest total background O_3; for the East, the period March-June appears to be when the total highest background levels occur. The previous conclusion in the 2014 PA (EPA, 2014a) and the 2013 ISA (EPA, 2013, in section 3.4) was that background O_3 was greatest over the U.S. during the spring and early summer (i.e., March-June), which agrees with the Jaffe et al. (2018) conclusion. The different patterns in the West noted in the PA (EPA, 2020b) do not agree with the patterns described by Dolwick et al. (2015) and Lefohn et al. (2014). The difference may be attributable to the lack of bias adjustment in the EPA model described in the current PA. The PA (EPA, 2020b) noted that bias adjustment was not performed in the modeling described in the PA. Lefohn et al. (2014) and Dolwick et al. (2015) performed bias adjustments. In their analyses, Lefohn et al. (2014) noted that model performance at low-elevation sites tended toward larger under prediction biases in the cool months (i.e., November-April) and larger over prediction biases in warm months (June-October), particularly for sites in the southern and eastern U.S. As is recognized today, hourly average concentrations associated with background O_3 can, at limited times and locations, be significantly higher as a result of stratospheric-tropospheric transport to the surface (Lefohn et al., 2011, 2012, 2014; Emery et al., 2012; Lin et al., 2012; Federal Register, 2015; EPA, 2020b). At many sites, stratospheric-tropospheric transport to the surface is important during the springtime. As noted in the PA (EPA, 2020b, page 2-66), the background O_3 modeling analysis did not attempt to quantify the contributions from individual Natural sources (e.g., lightning, soil, fires, stratosphere) or to address exceptional events beyond basic screening to remove very large fire plumes. The inability to adequately quantify the contribution of stratospheric-tropospheric transport to the surface for background O_3 may explain some of the discrepancy between the most current EPA background O_3 modeling results and those reported in the 2014 PA (EPA, 2014a).

Figure ES-6. Annual time series of regional average predicted MDA8 total O_3 concentration and contributions of each source (see legend) for the West (top), and the East (bottom). Natural is global natural sources, Intl is international anthropogenic sources, USA is U.S. anthropogenic sources, and Res-Anth is the residual anthropogenic. (Source: EPA, 2020b Fig. 2-23, page 2-49).

22. Empirical data indicate that as emission reductions occur across the U.S., the higher MDA8 concentrations shift at many O_3 monitoring sites from the summer toward the March-June months. In addition, as emission reductions occur, the distribution of hourly average concentrations shift from the higher values toward the middle values and the lower values shift upward toward the middle values. There is a compression of the distribution of hourly average O_3 concentrations. In addition, as emission reductions occur, background O_3 concentrations increase their percentage in the observed total O_3 concentration with the result that the compressed distribution of hourly average concentrations based on empirical data begins to resemble at some locations the distribution of background O_3. The patterns derived from empirical data showing the (1) compression of the distribution of hourly average O_3 concentrations and (2) seasonal shift from the summer months to the March-June period that result from emission reductions provide an opportunity to assess the adequacy of models that estimate background O_3 levels. Background O_3 is an important component of the margin of safety determinations. (Section 3).

23. The ISA (EPA, 2020a) attributes increasing trend patterns observed at high-elevation western U.S. sites to long-range transport from Asia. Long-range transport from Asia has not influenced trend patterns at all western U.S. 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 have not experienced increasing trends over the period 2000-2014. (Section 3.2.6).

24. 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-7 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% across all ranges of concentrations. In comparison, Fig. ES-8 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).

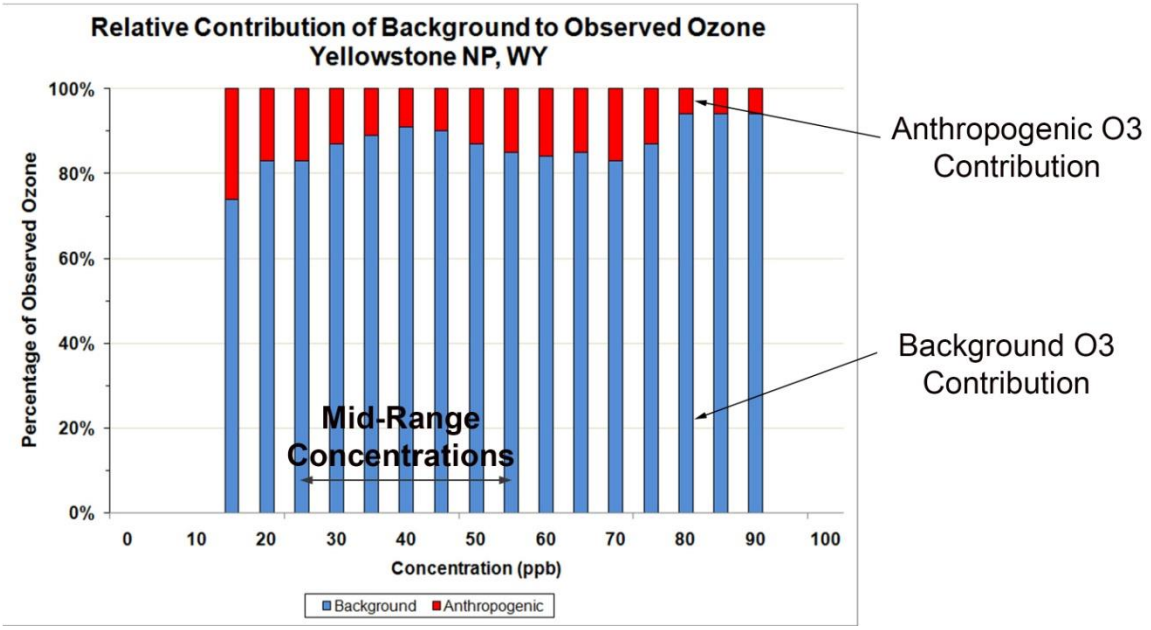


Figure ES-7. Average relative contributions of current hourly background (blue) and anthropogenic O_3 (red) for Yellowstone NP (WY) (AQS ID 560391011) in 2006. (Source: Lefohn et al., 2014).

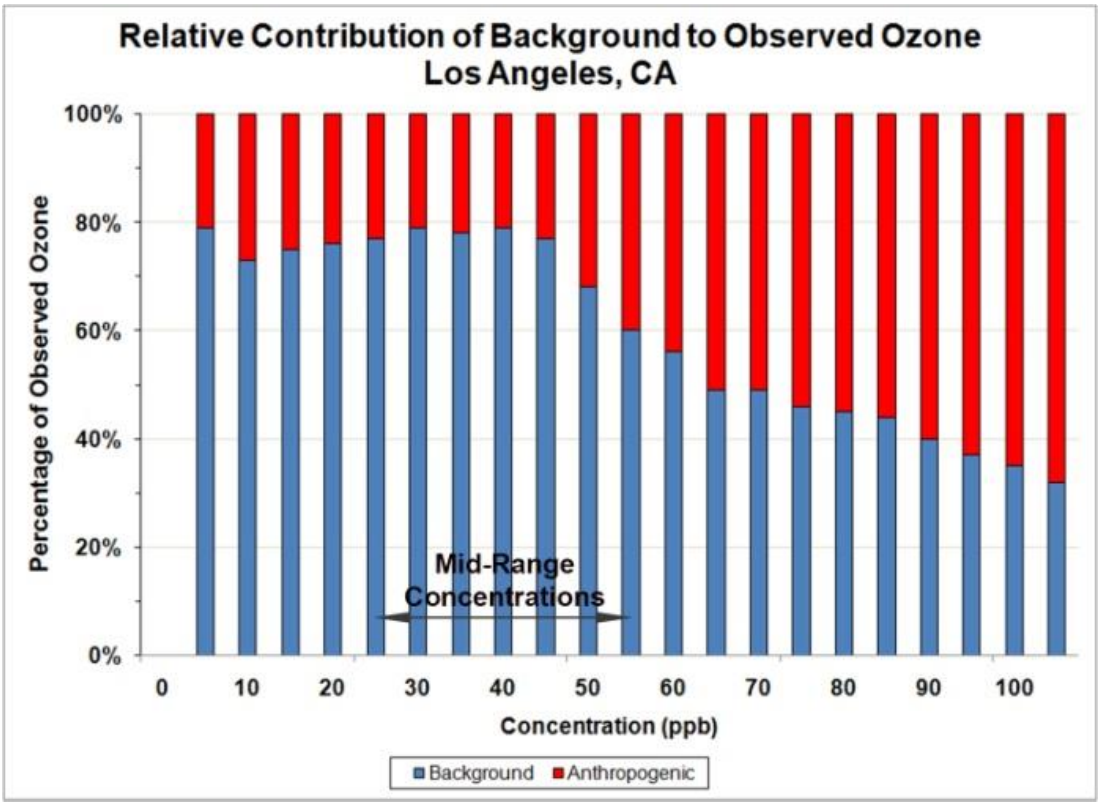


Figure ES-8. Average relative contributions of current hourly background (blue) and anthropogenic O_3 (red) for the Los Angeles (CA) (AQS ID 060719004) area in 2006. (Source: Lefohn et al., 2014).

25. In the PA (EPA, 2020b), analyses are presented that estimate exposure and risk for simulated populations in eight study areas (Atlanta, Boston, Dallas, Detroit, Philadelphia, Phoenix, Sacramento, and St. Louis). The eight study areas represent a variety of circumstances about population exposure to short-term concentrations of O_3 in ambient air. The eight study areas range in total population size from approximately two to eight million and are distributed across the U.S. in seven different NOAA climate regions: The Northeast, Southeast, Central, East North Central, South, Southwest and West. The PA did not provide examples of the time series for 2016 for the observed and USB concentrations for the eight sites used in the PA risk assessment. In Figs. ES-9 through ES-15, total observed O_3 concentrations, USB_{AB} estimates (data provided by the EPA), and STT-S counts are presented for 2007 for seven of the eight sites (i.e., Atlanta, Boston, Dallas, Detroit, Philadelphia, Sacramento, and St. Louis) used by the EPA in its risk analyses presented in the PA. In the figures, gaps (i.e., the difference between the observed total O_3 (noted by the black line) and USB_{AB} concentrations (noted by the green line)) occur, indicating the apparent influence of anthropogenic sources. (Section 3.2.9).

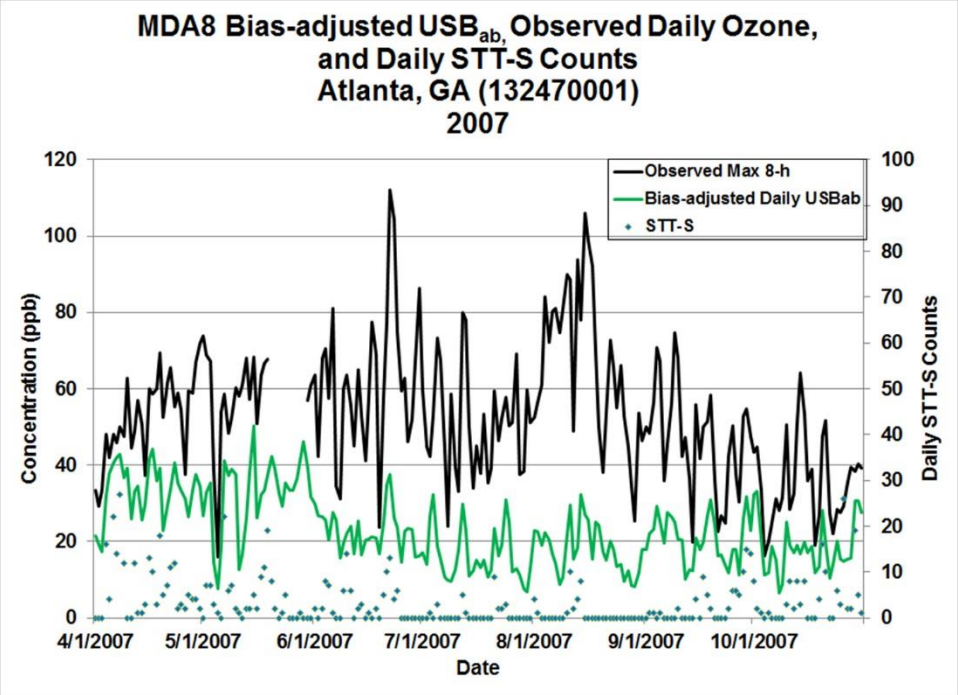


Figure ES-9. A comparison of the observed 8-h daily maximum concentration with the estimated bias-adjusted U.S. Background (USB_{AB}) 8-h daily maximum concentrations for a site in Atlanta, Georgia (AQS ID 132470001) for April-October 2007. The daily stratospheric-tropospheric transport to surface (STT-S) 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.

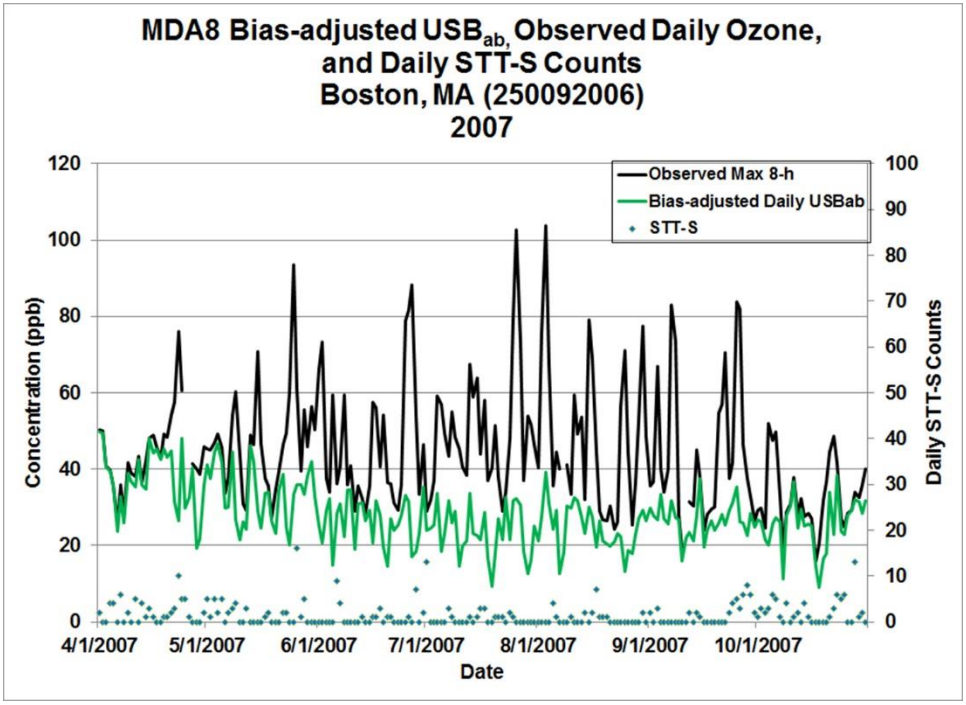


Figure ES-10. A comparison of the observed 8-h daily maximum concentration with the estimated bias-adjusted U.S. Background (USB_{AB}) 8-h daily maximum concentrations for a site in Boston, Massachusetts (AQS ID 250092006) for April-October 2007. The daily stratospheric-tropospheric transport to surface (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.

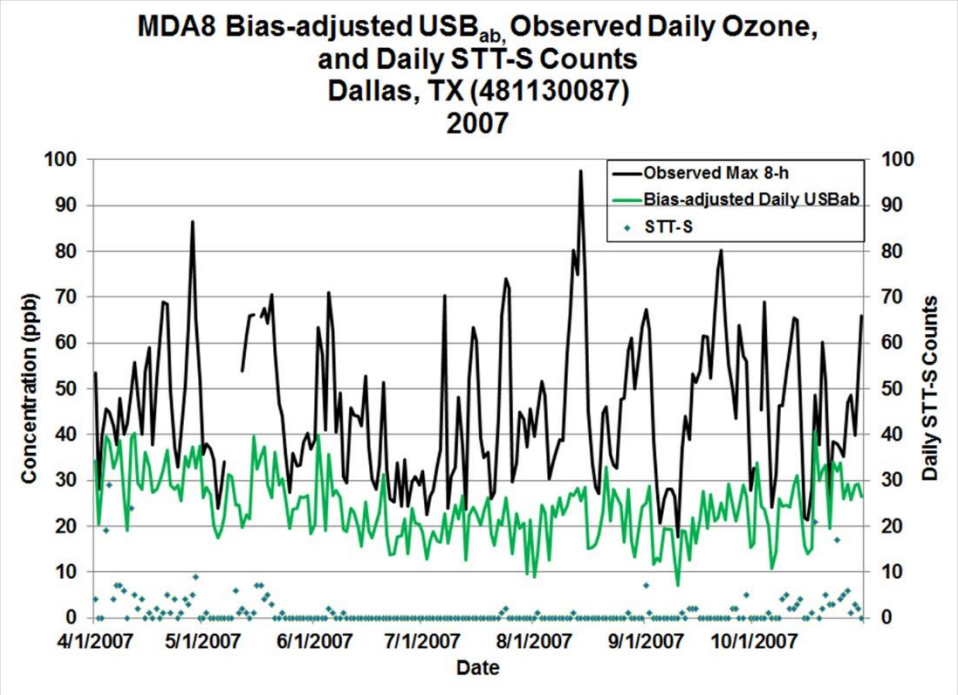


Figure ES-11. A comparison of the observed 8-h daily maximum concentration with the estimated bias-adjusted U.S. Background (USB_{AB}) 8-h daily maximum concentrations for a site in Dallas, Texas (AQS ID 481130087) for April-October 2007. The daily stratospheric-tropospheric transport to surface (STT-S) 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.

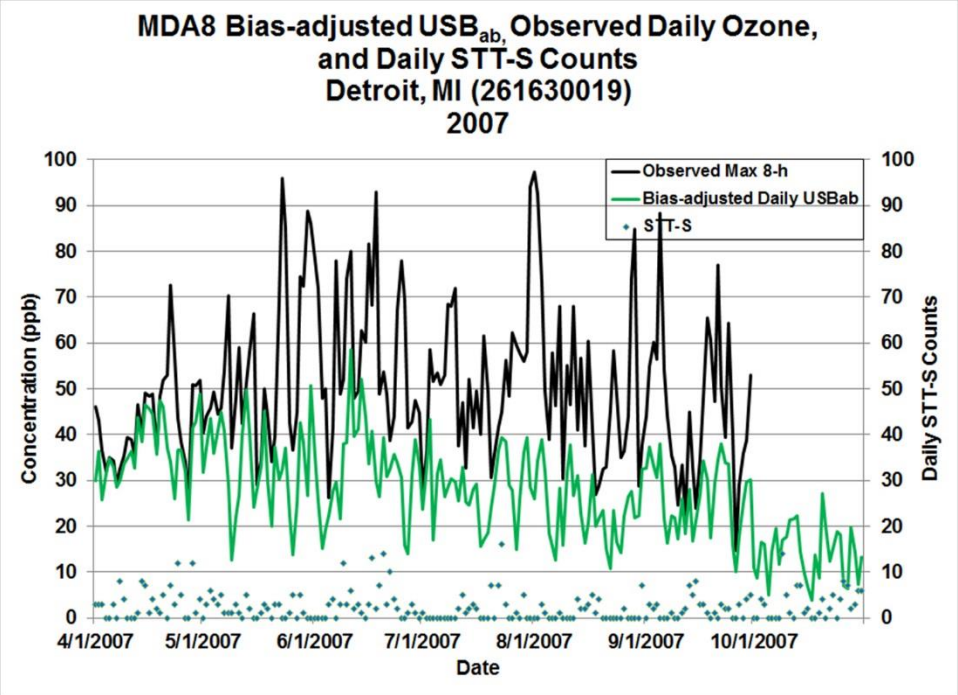


Figure ES-12. A comparison of the observed 8-h daily maximum concentration with the estimated bias-adjusted U.S. Background (USB_{AB}) 8-h daily maximum concentrations for a site in Detroit, Michigan (AQS ID 261630019) for April-October 2007. The daily stratospheric-tropospheric transport to surface (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.

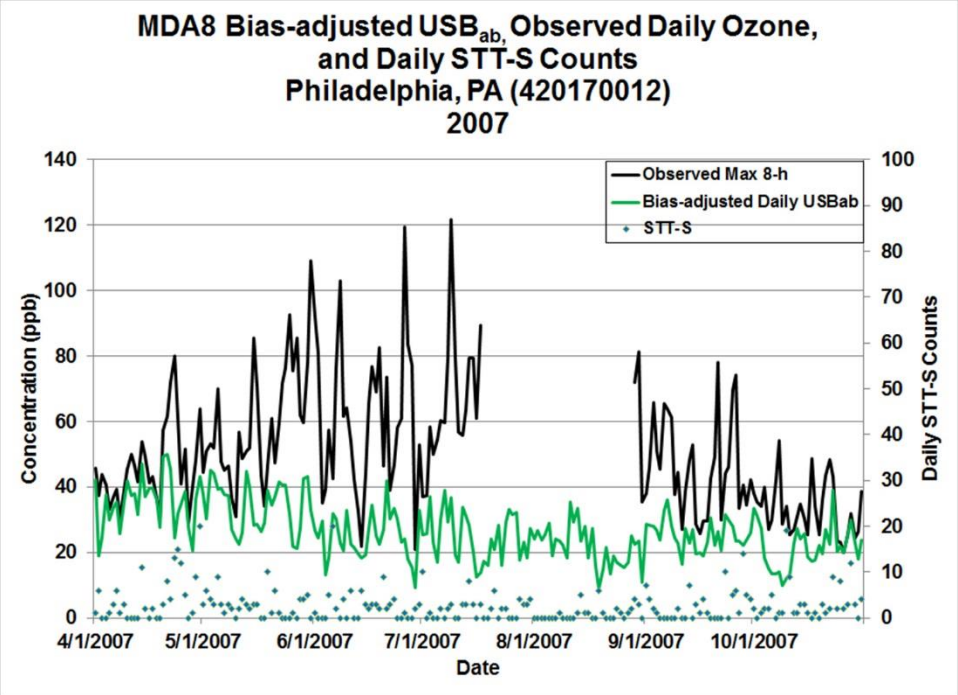


Figure ES-13. A comparison of the observed 8-h daily maximum concentration with the estimated bias-adjusted U.S. Background (USB_{AB}) 8-h daily maximum concentrations for a site in Philadelphia, Pennsylvania (AQS ID 420170012) for April-October 2007. The daily stratospheric-tropospheric transport to 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.

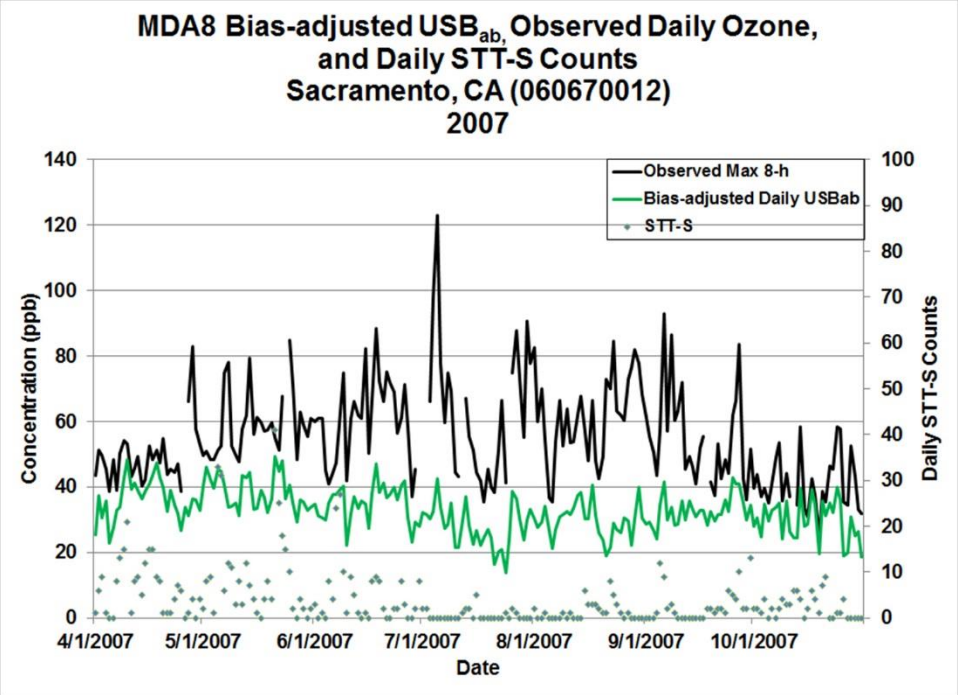


Figure ES-14. A comparison of the observed 8-h daily maximum concentration with the estimated bias-adjusted U.S. Background (USB_{AB}) 8-h daily maximum concentrations for a site in Sacramento, California (AQS ID 060670012) for April-October 2007. The daily stratospheric-tropospheric transport to surface (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.

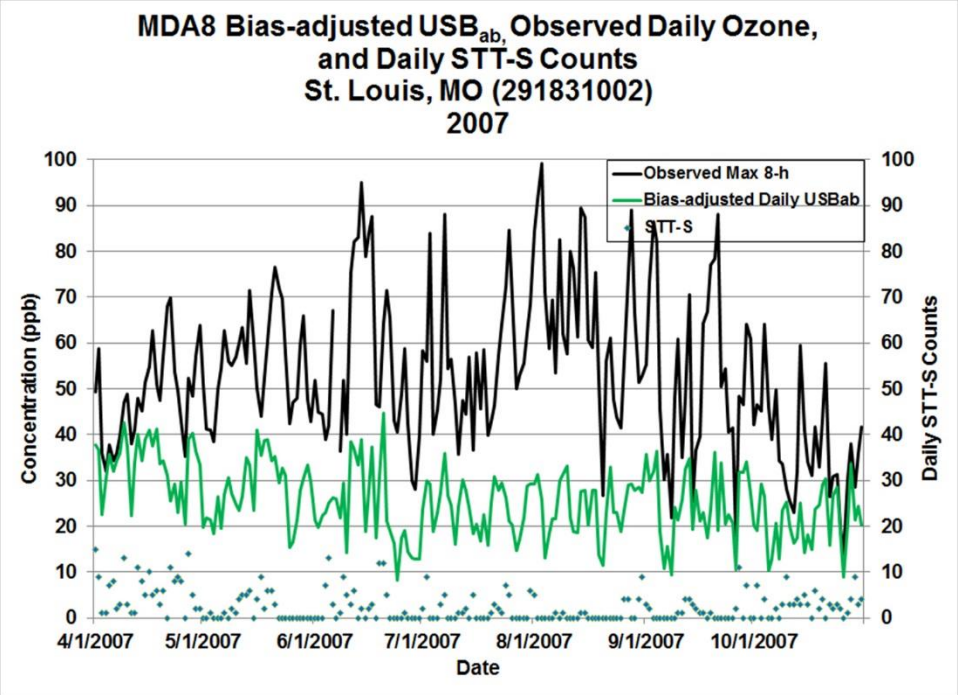


Figure ES-15. A comparison of the observed 8-h daily maximum concentration with the estimated bias-adjusted U.S. Background (USB_{AB}) 8-h daily maximum concentrations for a site in St. Louis, Missouri (AQS ID 291831002) for April-October 2007. The daily stratospheric-tropospheric transport to surface (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.

1. Introductory Comments

This Draft Review of the Ozone National Ambient Air Quality Standards document (Federal Register, 2020) presents the Administrator's proposed decisions in the current review of the primary (health-based) and secondary (welfare-based) O_3 NAAQS. In so doing, this document summarizes the background and rationale for the Administrator's proposed decisions to retain the current standards, without revision. In reaching his proposed decisions, the Administrator has considered the currently available scientific evidence in the Integrated Science Assessment (ISA) document (EPA, 2020a), quantitative and policy analyses presented in the Policy Assessment (PA) document (EPA, 2020b), and advice from the Clean Air Scientific Advisory Committee (CASAC).

The last review of the O_3 NAAQS, completed in 2015, established the current primary and secondary standards (80 FR 65291, October 26, 2015). In the decision on subsequent litigation on the 2015 decisions, the U.S. Court of Appeals for the District of Columbia Circuit (DC Circuit) upheld the 2015 primary standard but remanded the 2015 secondary standard to the EPA for further justification or reconsideration (US Court of Appeals, DC Circuit, August 23, 2019). As EPA notes in its current draft document, the Court's remand of the secondary standard was considered in reaching the proposed decision, and the associated proposed conclusions and judgments, described in its Draft Review of the Ozone National Ambient Air Quality Standards document. In Section 1.9, I have provided comments on the EPA's rationale for using the current form of the O_3 NAAQS as a substitute for the biologically relevant W126 exposure metric.

1.1 Purpose of the Integrated Science Assessment (ISA) Document

The purpose of the 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 was 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 (EPA, 2006), the 2007 Staff Paper (EPA, 2007), the 1996 AQCD and

Staff Paper for Ozone and Other Photochemical Oxidants (EPA, 1996a, b), the 1986 AQCD for Ozone (EPA, 1986) and its Supplement (EPA, 1992), and the 1978 AQCD for Ozone and Other Photochemical Oxidants (EPA, 1978)). It was 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 was important was that the latest results were compared to past findings and where disagreement was identified, space was provided in the ISA to discuss the ramifications of the disagreement. In some cases, the final version of the ISA did not focus on scientific disagreements and did not critically evaluate possible reasons for the disagreement in the results. The failure to critically evaluate the disagreements was not helpful in reducing uncertainties associated with margin of safety considerations. I will discuss specific examples later in my comments.

As indicated above, in reaching his proposed decisions on the two Federal O_3 standards, the Administrator considered as part of his current draft recommendation the currently available scientific evidence in the Integrated Science Assessment (ISA) document (EPA, 2020a). In his April 1, 2020 letter to Dr. Louis Anthony Cox, Jr., Chair of CASAC, the Administrator (EPA, 2020c) noted that

> The CASAC has raised a number of important issues with the draft Ozone ISA. In addition to offering numerous consensus and individual comments on scientific issues, the CASAC has requested that the agency incorporate substantial changes to our documentation procedures. As you noted in your letter, the CASAC finds that the draft Ozone ISA "does not provide a comprehensive, systematic assessment of the available science relevant to understanding the public health impacts of changes in ambient concentrations of ozone." **Some of these comments and adjustments can be addressed in the near term, while others will require additional time to complete** (emphasis added).

The Administrator continues as follows in his letter to CASAC:

> The process outlined in the EPA's May 9, 2018, "Back-to-Basics" memo directs the agency to ensure that NAAQS reviews are completed in a timely, efficient and transparent manner, consistent with the *Clean Air Act.* The five-year review cycle for each NAAQS is challenging in light of the continuous development of new and relevant science, challenges compounded by the EPA practice of facilitating CASAC and public engagement throughout the process. With this in mind, I have directed my staff to do the following:
>
> - Complete the review of the Ozone NAAQS by the end of 2020. The difficulty of this task is not lost on me, and I recognize that the CASAC has raised concerns regarding the limitations of the current draft Ozone ISA. I have asked that staff maintain their focus on meeting our statutory deadlines while reflecting the latest scientific information in final Ozone ISA. In practical terms, this means:

- By April 2020, incorporate the CASAC's comments and recommendations, to the extent possible, and create a final Ozone ISA so that it may be available to inform a proposed decision on any necessary revisions of the NAAQS by the spring of 2020.

- Based on initial feedback from the CASAC and while making necessary adjustments to the draft Ozone ISA, keep production of the Ozone Policy Assessment, including relevant exposure and risk assessment work, on schedule to create a final Ozone Policy Assessment by the spring of 2020.

- For those comments and recommendations that are more substantial or crosscutting and which cannot be fully addressed in this timeframe, develop a plan to incorporate these changes in future Ozone ISAs as well as ISAs for other criteria pollutant reviews.

The Administrator concludes in his April 1, 2020 letter to Dr. Cox, Chair of CASAC

> I believe that sound science must be the foundation upon which all the EPA's regulatory and policy decisions are based. Independent reviews such as those of the CASAC help ensure that the agency uses the best available science to fulfill its mission to protect human health and the environment.

The EPA Administrator indicated in his letter (EPA, 2020c) that the final version of the Ozone ISA (EPA, 2020a) would not necessarily reflect a complete response to the concerns of the CASAC. Therefore, it may be possible that the final version of the ISA (EPA, 2020a) did not provide a comprehensive, systematic assessment of the available science relevant to understanding the public health and environmental impacts of changes in ambient O_3 concentrations.

1.2 Purpose of the Policy Assessment (PA) Document

As indicated in the PA (EPA, 2020b), "The PA, when final, presents an evaluation, for consideration by the EPA Administrator, of the policy implications of the currently available scientific information, assessed in the ISA, any quantitative air quality, exposure or risk analyses based on the ISA findings, and related limitations and uncertainties." The role of the PA is to assist in the bridging of the gap between the Agency's scientific assessment and quantitative technical analyses, and the judgments required of the Administrator in determining whether it is appropriate to retain or revise the NAAQS.

The development of the PA (EPA, 2020b) is also intended to facilitate advice to the Agency and recommendations to the Administrator from an independent scientific review committee, the Clean Air Scientific Advisory Committee (CASAC), as provided for in the Clean Air Act (CAA). The CASAC is to advise on subjects including the Agency's assessment of the relevant scientific information and on the adequacy of the current standards, and to make

recommendations as to any revisions of the standards that may be appropriate. In the past, the EPA generally made available to the CASAC and the public one or more drafts of the PA (EPA, 2020b) for CASAC review and public comment. In the current rulemaking activity, only one draft of both documents was made available for review.

Beyond informing the Administrator and facilitating the advice and recommendations of the CASAC, the PA (EPA, 2020b) is also intended to be a useful reference to all parties interested in the review of the O_3 NAAQS. In these roles, it is intended to serve as a source of policy-relevant information that supports the Agency's review of the O_3 NAAQS, and it is written to be understandable to a broad audience.

An important challenge of the PA (EPA, 2020b) is how to integrate the results in the current PA with those results from the previous Ozone PA (EPA, 2014a) and place all of these results into perspective with key findings contained within previous O_3 rulemaking documents (e.g., 2006 AQCD for Ozone and Related Photochemical Oxidants (EPA, 2006), the 2007 Staff Paper (EPA, 2007), the 1996 AQCD and Staff Paper for Ozone and Other Photochemical Oxidants (EPA, 1996a, b), the 1986 AQCD for ozone (U.S. EPA, 1986) and its Supplement (EPA, 1992), and the 1978 AQCD for Ozone and Other Photochemical Oxidants (EPA, 1978)). It is extremely important that the PA 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 PA devotes space to discussing the ramifications of the disagreement. In some cases, the PA did not focus sufficiently on the disagreement with past results and did not critically evaluate possible scientific reasons for the disagreement. Specific examples are discussed in my 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. The compressed schedule during this rulemaking cycle resulted in the simultaneous preparation of the draft ISA (EPA, 2019a) and draft PA (EPA, 2019b) documents. In contrast, during the last O_3 rulemaking activities, additional time was permitted for the review and accurate integration of the various rulemaking documents. Previously, 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. On December 2, 2019, I submitted my comments on the draft Integrated Scientific Assessment Document (Lefohn, 2019a) to the Docket. On December 16, 2019, I submitted to the Docket my comments on the draft of the EPA Policy Assessment for the Review of the Ozone National Ambient Air Quality Standards (Lefohn, 2019b). Clearly, the adequacy of the scientific integrity of the PA document (EPA, 2020b) is influenced by the accuracy of the state-of-science summary contained within the ISA document (EPA, 2020a). As noted in his April 1, 2020 letter to CASAC (EPA, 2020c), because of time limitations, the final version of the ISA may not necessarily reflect the latest state-of-science evidence. The inadequacies contained within both the ISA and PA, which are

reflected in the Administrator's draft document (Federal Register, 2020), are discussed in my comments.

1.3 Important Concepts Guiding the Human Health and Welfare Ozone Standards

There are two key scientific fundamental principles that help guide the form and level of the health and welfare Federal O_3 standards in the United States (see Lefohn, 2019a and Lefohn, 2019b for a detailed explanation). 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**.

Without adhering to the first fundamental principle, the selection of O_3 exposure indices for health and welfare assessment purposes would be based on the personal choice of the investigator rather than based on biologically relevant metrics developed under laboratory or empirical conditions. Lefohn et al. (2018), in a 24-coauthor international Tropospheric Ozone Assessment Report (TOAR), discussed the rationale for the selection of specific exposure metrics for assessing human health and vegetation. The first fundamental principle provides guidance to policymakers and researchers on which part of the distribution of hourly average O_3 concentrations are biologically relevant for assessing human health and vegetation effects.

As noted in the Administrator's Draft Review of the Ozone National Ambient Air Quality Standards document (Federal Register, 2020 – page 49842), the EPA Administrator, in her 2015 decision, set the human health standard at 70 ppb. The decision was based on the following rationale:

> The 2015 decision to set the level of the revised primary O_3 standard at 70 ppb built upon the Administrator's conclusion (summarized in section II.A.1.a above) that the overall body of scientific evidence and exposure/risk information called into question the adequacy of the public health protection afforded by the then-current standard, particularly for at-risk populations and lifestages (80 FR 65362, October 26, 2015). In her decision on level, the Administrator placed the greatest weight on the results of controlled human exposure studies and on quantitative analyses based on information from these studies, particularly analyses of O_3 exposures of concern. In so doing, the Administrator noted that controlled human exposure studies provide the most certain evidence indicating the occurrence of health effects in humans following specific O_3 exposures, noting in particular that the effects reported in the controlled human exposure studies are due solely to O_3 exposures, and are not complicated by the presence of co-occurring pollutants or pollutant mixtures (as is the case in epidemiologic studies).

In controlled human health clinical studies (Hazucha et al., 1992; Adams 2003, 2006a, b), greater O_3 hour-by-hour peak responses were observed in stepwise and triangular (i.e., smooth increases and decreases in concentration) exposures rather than in constant concentration exposure protocols. Because of the (1) greater importance of the higher O_3 concentrations and

(2) behavior of the metric as emissions are reduced, annual, seasonal, or monthly average concentrations may not be appropriate exposure metrics to use for estimating either acute or chronic human health effects (see Section 1.4). The focus by the EPA on the higher part of the distribution of the hourly average O_3 concentrations rather than the lower part provides clear guidance to those who are responsible for developing emissions reduction strategies to protect the public.

EPA's recognition of the importance of the higher O_3 concentrations is reflected in its use of the Air Quality Index (AQI) reported across the U.S. (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 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. 1-1 below. The higher the O_3 concentration exposures, the greater the potential effect on human health.

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 1-1. Air quality index levels (AQI) related to 8-h concentrations.

For vegetation, the EPA ISA (2013, 2020a) concluded 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. Although not mentioned in Federal Register (2020), results from a "natural experiment" site in the San Bernardino National Forest, where substantial reductions over the years in the higher hourly average O_3 concentrations in the Los Angeles area occurred, provides independent confirmation of the experimental studies described below for the greater importance of the higher hourly average O_3 concentrations in influencing vegetation effects. The 2013 ISA (EPA, 2013) 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 O_3 concentrations was little changed over this period. EPA (2013) suggested it was the reduction in the higher hourly average O_3 concentrations that was responsible for the improvement in tree health.

The key vegetation experimental studies that support the importance of the higher O_3 concentrations were performed 30 to 40 years ago (i.e., mostly in the 1980s and 1990s). Musselman et al. (1983) and Hogsett et al. (1985) performed research experiments that showed that the higher hourly average concentrations were more important than the mid- and low-level values in eliciting adverse vegetation effects. Following their results, a series of controlled experiments was undertaken worldwide for assessing the importance of the higher O_3 concentrations in eliciting vegetation response. These controlled fumigation experimental results (e.g., EPA, 1986, 1992, 1996a, 1996b, 2013; Musselman et al., 1986, 1994; Nussbaum et al., 1995; Yun and Laurence, 1999; Lee and Hogsett, 1999; Oksanen and Holopaninen, 2001; Köllner and Krause, 2003; Wang et al., 2008) provided additional evidence supporting the findings by Musselman et al. (1983) and Hogsett et al. (1985) that higher concentrations should receive greater weighting in comparison to the mid- and low-level values.

The first fundamental principle indicates that for human health and vegetation effects purposes, the higher hourly average O_3 concentrations should be provided greater attention than the mid- and lower values within the distribution of hourly average concentrations. The second fundamental principle addresses what happens within the distribution of hourly average concentrations when emissions reduction occurs to protect human health and welfare. The second fundamental principle is

> **Daily Maximum Hourly Averaged O_3 Concentrations Will Remain Well above 0 Parts per Billion (ppb) Even if all Anthropogenic Emissions Were Eliminated Worldwide**.

As O_3 precursor emissions are reduced at specific locations across the U.S. to attain the current O_3 NAAQS, not all the hourly average O_3 concentrations at these locations shift downward. Empirical data and atmospheric chemistry/meteorological modeling results indicate that as emissions are reduced at many sites across the U.S., a compression of the higher and lower hourly average O_3 concentrations toward the middle of the distribution occurs. For example, as emissions reduction occurs, using the 8-h daily maximum (MDA8) concentrations, the MDA8 values are reduced downward toward the mid-level concentrations and the lowest MDA8 values ***increase***. The shift is associated with less titration of O_3 by NO of the lower

hourly average concentrations as reduction in NO_x emissions occur (Lefohn et al., 1998; EPA, 2014b; Simon, 2015; Lefohn et al., 2017, 2018; EPA, 2020b).

The compression of the distribution of hourly average O_3 concentrations as noted above results in annual average or median concentration values ***increasing*** at some sites. For example, in Fig. 1-2 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 8-h O_3 exposures that are lower than the two urban sites. When the annual average is plotted (Fig. 1-3) for the same time with the same hourly averaged 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 the ordering when the annual average index is used. The increase in the annual average concentration values, even though emissions are reduced, is associated with the low end of the distribution increasing due to less titration of O_3 by NO as NO_x emissions are reduced The three National Park sites in the figures do not experience high 8-h average daily maximum concentration values comparable to many of the urban sites in the U.S. The high-elevation Yellowstone National Park site experiences much higher annual average values than 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). No trend has been observed at Yellowstone National Park using the 4th highest daily maximum 8-h average concentration metric when nonparametric statistics are applied.

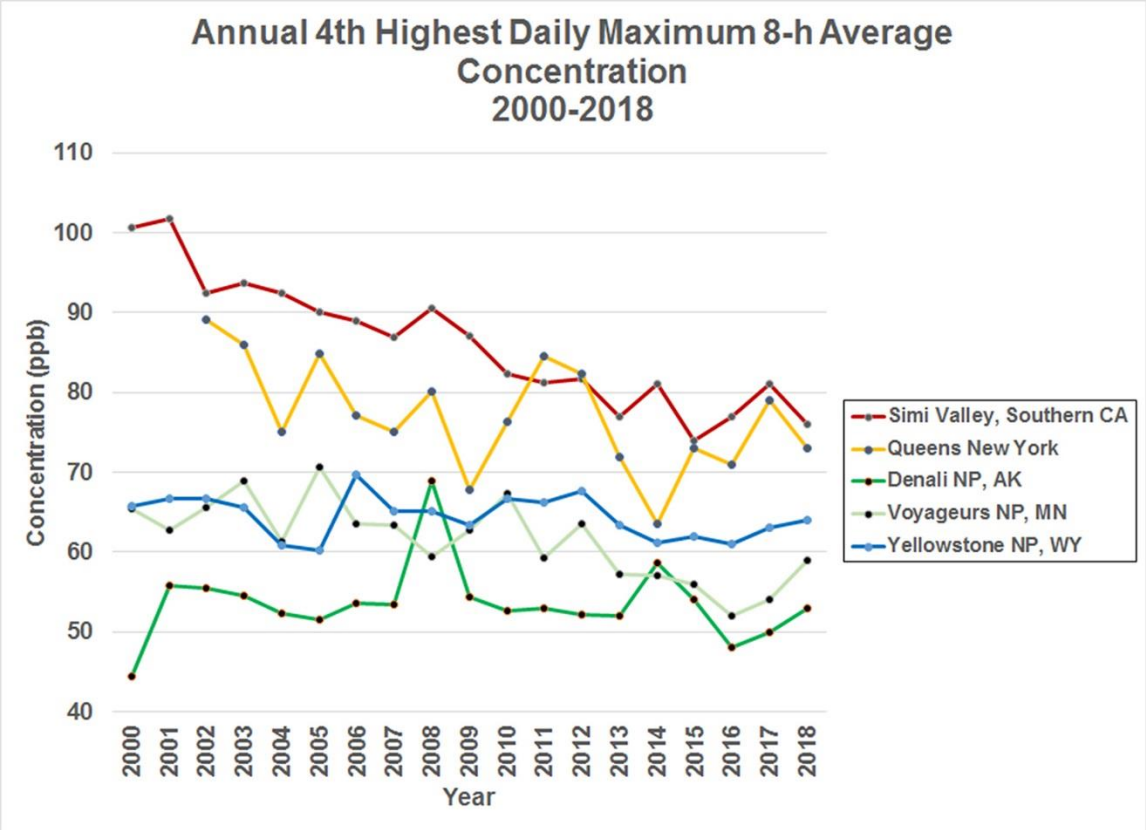


Figure 1-2. The annual 4th highest daily maximum 8-h average O_3 concentration for the period 2000-2018 for Simi Valley, CA (061112002), Queens New York, NY (360810124, Denali National Park, AK (020680003), Voyageurs National Park, MN (271370034), and Yellowstone National Park, WY (560391011). Source of data is from the EPA's AQS database.

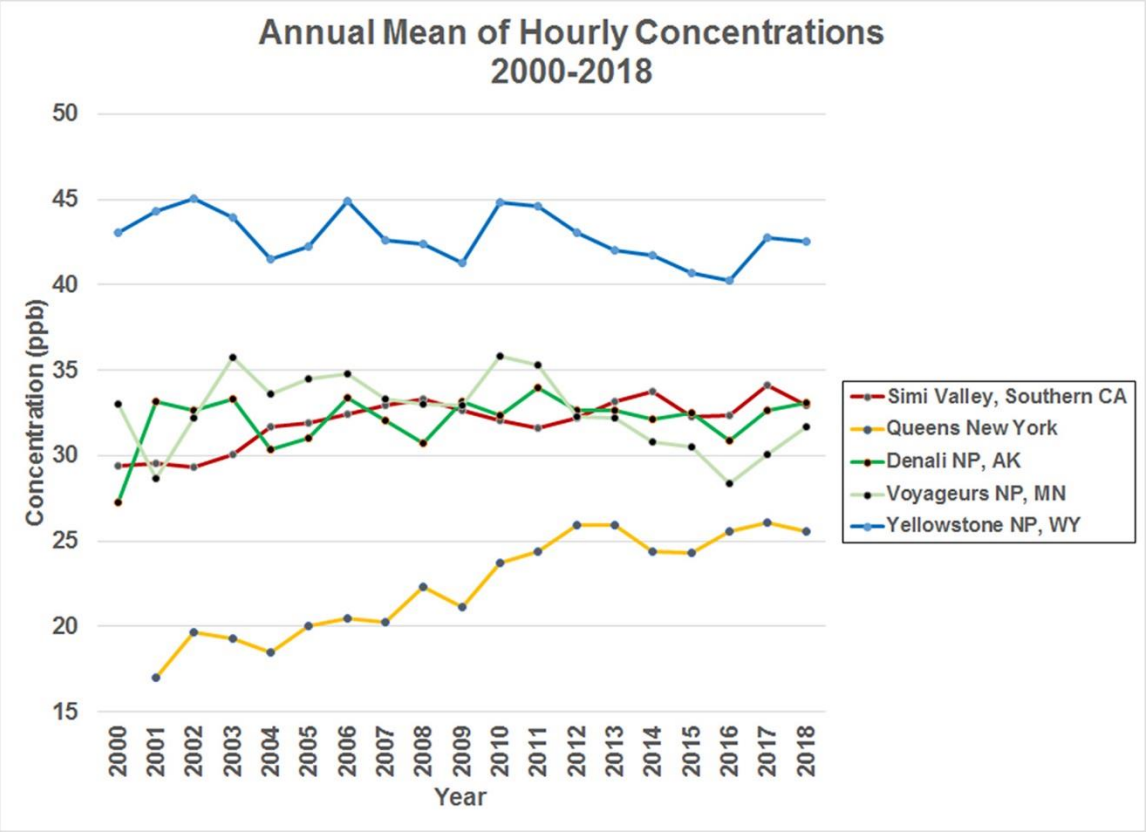


Figure 1-3. The annual mean of the hourly average O_3 concentrations for the period 2000-2018 for Simi Valley, CA (061112002), Queens New York, NY (360810124, Denali National Park, AK (020680003), Voyageurs National Park, MN (271370034), and Yellowstone National Park, WY (560391011). Source of data is from the EPA's AQS database.

Besides annual averages increasing as emissions are reduced, the current PA (EPA, 2020b) notes that metrics impacted by averaging over longer time periods of hourly O_3 measurements, such as the 6-month (April-September) average of daytime (8am-7pm) O_3 concentrations, were more varied with only about half of the sites exhibiting decreases in this metric and most other sites exhibiting no trend (Lefohn et al., 2017) as emissions were reduced. Thus, long-term average exposure metrics appear to have serious limitations for assessing risks associated with O_3 exposures. The inconsistency of trends associated with the use of annual average and other long average metrics is linked to the titration of O_3 by NO as NO_x emissions are reduced to protect the public's health and welfare. The inconsistency affects the efficacy of the use of long-term metrics, such as the annual average and 6-month average concentrations, for assessing human health and vegetation effects and risks.

1.4 Acute and Chronic Ozone Effects

The Administrator noted (Federal Register, 2020 - page 49842) that in the Agency's 2015 review of the O_3 NAAQS, that

> In the review completed in 2015, the Administrator concluded, in consideration of the then-available health effects information, that an 8-hour averaging time remained appropriate for addressing health effects associated with short-term exposures to ambient air O_3 and that it could effectively limit health effects attributable to both short- and long-term O_3 exposures (80 FR 65348, October 26, 2015).

In its review of the 2015 O_3 NAAQS rulemaking (Federal Register, 2015 – page 65358), the Administrator noted:

> In considering estimates of exposures of concern for the 60, 70, and 80 ppb benchmarks within the context of her judgments on adversity, the Administrator notes that, due to interindividual variability in responsiveness, not every occurrence of an exposure of concern will result in an adverse effect. As discussed above (II.B.2.b.i), this point was highlighted by some commenters who opposed revision of the current standard, based on their analysis of effects shown to occur following exposures to 72 ppb O_3. 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).

Thus, in the Administrator's decision in 2015, the Agency believed 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 O_3 average concentrations shifts downwards toward the middle part of the hourly average concentration distribution.

1.5 Margin of Safety Considerations

As noted in the Administrator's Draft Review of the Ozone National Ambient Air Quality Standards document (Federal Register, 2020 – page 49833):

> Section 109 [42 U.S.C. 7409] directs the Administrator to propose and promulgate "primary" and "secondary" NAAQS for pollutants for which air quality criteria are issued [42 U.S.C. 7409(a)]. Section 109(b)(1) defines primary standards as ones "the attainment and maintenance of which in the judgment of

> the Administrator, based on such criteria and allowing an adequate margin of safety, are requisite to protect the public health."

The requirement that primary standards provide an adequate margin of safety was intended to address uncertainties associated with inconclusive scientific and technical information available at the time of standard setting (Federal Register, 2020 – page 49833). The margin of safety was also intended to provide a reasonable degree of protection against hazards that research has not yet identified. As noted by the Administrator (Federal Register, 2020 - page 49833):

> Thus, in selecting primary standards that include an adequate margin of safety, the Administrator is seeking not only to prevent pollution levels that have been demonstrated to be harmful but also to prevent lower pollutant levels that may pose an unacceptable risk of harm, even if the risk is not precisely identified as to nature or degree. The CAA does not require the Administrator to establish a primary NAAQS at a zero-risk level or at background concentration levels (see *Lead Industries Ass'n v. EPA*, 647 F.2d at 1156 n.51, *Mississippi v. EPA*, 744 F.3d at 1351), but rather at a level that reduces risk sufficiently so as to protect public health with an adequate margin of safety.
>
> In addressing the requirement for an adequate margin of safety, the EPA considers such factors as the nature and severity of the health effects involved, the size of the sensitive population(s), and the kind and degree of uncertainties. The selection of any particular approach to providing an adequate margin of safety is a policy choice left specifically to the Administrator's judgment. See Lead Industries Ass'n v. EPA, 647 F.2d at 1161-62; Mississippi v. EPA, 744 F.3d at 1353.

In NAAQS reviews generally, evaluations of how particular primary standards address the requirement to provide an adequate margin of safety include consideration of such factors as the nature and severity of the health effects, the size of the sensitive population(s) at risk, and the kind and degree of the uncertainties present. The Administrator noted that in the 2015 decision, given (1) the consideration of the evidence, (2) exposure and risk information, (3) advice from the CASAC, and (4) public comments, judged that a revised primary standard of 70 ppb, in terms of the 3-year average of the annual 4th highest daily maximum 8-hour average O_3 concentrations, was sufficient to protect public health, including the health of at-risk populations, with an adequate margin of safety (Federal Register, 2020 - page 49843).

As noted above, the decision process for deciding an adequate margin of safety involves consideration of such factors as the nature and severity of the health effects, the size of the sensitive population(s) at risk, and the kind and degree of the uncertainties. One factor that influences the degree of uncertainties is background O_3. According to a Court decision in 2019, the current rulemaking background O_3 should not directly influence the setting of the level of the NAAQS. The Administrator notes (Federal Register, 2020 - page 49836) that in the decision on subsequent litigation on the 2015 O_3 NAAQS decisions, the U.S. Court of Appeals for the District of Columbia Circuit (DC Circuit) in its August 19, 2019 decision addressed arguments

regarding considerations of background O_3 concentrations, and socioeconomic and energy impacts. Regarding background O_3, the Court rejected the argument that the EPA was required to take background O_3 concentrations into account when setting the NAAQS. The Court found that the text of the Clean Air Act 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 the EPA did not act unlawfully or arbitrarily or capriciously in setting the 2015 NAAQS without regard for background O_3.

While background O_3 currently, as per the Court's August 19, 2019 decision, is not a direct consideration in the setting of the level of the O_3 standard, background O_3 is an important consideration for assessing human health effects risks. The risk assessments play an important role in the margin of safety determinations and thus, background O_3, in an indirect manner, could influence the level of the O_3 standard. Background O_3 concentrations in the low- and mid-level part of the distribution of concentrations make up a large fraction of the total ambient O_3 levels and potentially can influence human health risk assessments associated with margin of safety determinations. The EPA notes that across the ensemble of available modeling studies in the literature, ***seasonal mean*** daily maximum 8-h background O_3 concentrations are estimated to range from 20−50 ppb (EPA, 2020a - page 1-1). This means that over an entire O_3 season, hourly average background O_3 concentration can be higher than the 20-50 ppb seasonal mean of the daily maximum 8-h range of values. In many cases, 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)), including the low-level values associated with background O_3. For lung function probabilistic population-based Exposure-Response (E-R) functions risk assessments, the lower concentrations, which are at background O_3 levels, have the potential to contribute to the total risk at low-elevation sites because of the more frequent occurrences than the higher values. As noted above, an adequate margin of safety is a policy choice left specifically to the Administrator's judgment. The greater the contribution of background O_3 to the human health risk assessment, the greater the uncertainty will be to the input into the margin of safety consideration. Thus, because of its importance in affecting the human health risk assessments used in the margin of safety determination, background O_3 should have been quantified for the 8 cities highlighted in the Agency's modeling analyses (i.e., Atlanta, Boston, Dallas, Detroit, Philadelphia, Phoenix, Sacramento, and St. Louis) in the PA (EPA, 2020b). However, quantification of background O_3 for the 8 cities was not performed. Therefore, no information was provided in the current O_3 NAAQS rulemaking process concerning the contribution of background O_3 to the human health effects risk assessments that provided valuable information on the margin of safety.

1.6 Quantifying Background Ozone for Margin of Safety Considerations

As noted above, the Court recognized that NAAQS O_3 levels are set to protect public health and welfare and that background O_3 is not a consideration in setting the primary (i.e.,) human health standard. As mentioned in the previous section, while background O_3 currently is not a consideration in the setting of the level of the O_3 standard, background is an important

consideration for assessing human health effects risks. The risk assessments play an important role in the margin of safety determinations. It is important to understand the concentration levels at which background O_3 plays important roles in influencing human health risk assessments.

The EPA generally characterizes O_3 concentrations that would exist in the absence of U.S. anthropogenic emissions as U.S. background. As indicated in the current PA (EPA, 2020b; page 2-4), sources of emissions of O_3 precursor compounds can be divided into anthropogenic and natural source categories, with natural sources further divided into emissions from biological processes of living organisms (e.g., plants, microbes, and animals) and emissions from chemical or physical processes (e.g., biomass burning, lightning, and geogenic sources). The current PA (EPA, 2020b; page 2-29) notes that natural sources are considered background regardless of where they occur. By contrast, anthropogenic sources are only considered as background when they are not from sources within the focus area. In the context of EPA's definition of background O_3, anthropogenic background is synonymous with O_3 originating from international anthropogenic emission sources. As noted in the current PA (EPA, 2020b), the relative contribution of international and natural background sources is most notably larger at locations near borders (i.e., international) or high-elevation (i.e., natural) locations.

In this context, the Administrator notes (Federal Register, 2020, pages 49837-49838) that

> Concentrations of O_3 in ambient air that result from natural and non-U.S. anthropogenic sources are collectively referred to as U.S. background O_3 (USB; PA, section 2.5). As in the last review, we generally characterize O_3 concentrations that would exist in the absence of U.S. anthropogenic emissions as U.S. background (USB). Findings from modeling analyses performed for this review to investigate patterns of USB in the U.S. are largely consistent with conclusions reached in the last review (PA, section 2.5.4). The current modeling analysis indicates spatial variation in USB O_3 that is related to geography, topography and proximity to international borders and is also influenced by seasonal variation, with long-range international anthropogenic transport contributions peaking in the spring while U.S. anthropogenic contributions tend to peak in summer. The West is predicted to have higher USB concentrations than the East, with higher contributions from natural and international anthropogenic sources that exert influences in western high-elevation and near-border areas. The modeling predicts that for both the West and the East, days with the highest 8-hour concentrations of O_3 generally occur in summer and are likely to have substantially greater concentrations due to U.S. anthropogenic sources. While the USB contributions to O_3 concentrations on days with the highest 8-hour concentrations are generally predicted to come largely from natural sources, the modeling also indicates that a small area near the Mexico border may receive appreciable contributions from a combination of natural and international anthropogenic sources on these days. In such locations, the modeling suggests the potential for episodic and relatively infrequent events with substantial background contributions where daily maximum 8-hour O_3 concentrations approach or exceed the level of the current NAAQS (*i.e.*, 70 ppb). This contrasts with most monitor locations in the U.S. for which international contributions are predicted to be the

lowest during the season with the most frequent occurrence of daily maximum 8-hour O_3 concentrations above 70 ppb. This is generally because, except for in near border areas, larger international contributions are associated with long distance transport and that is most efficient in the springtime (PA, section 2.5.4).

The EPA notes that across the ensemble of available modeling studies in the literature, ***seasonal mean*** daily maximum 8-h background O_3 concentrations are estimated to range from 20−50 ppb (EPA, 2020a, page 1-1). While the seasonal mean of background O_3 may be in the 20 – 50 ppb range, the maximum daily 8-h background O_3 concentration will be higher than the 20 – 50 ppb seasonal mean range. For comparison, the current O_3 NAAQS is 70 ppb (annual 4th highest MDA8 value averaged over a 3-year period). Fig. 1-4 below illustrates the Agency's latest modeling estimates of the annual time series of the regional averages (including both low- and high-elevation sites) predicted for daily maximum 8-h concentrations (MDA8) for the West and East. Background O_3 is higher in the western U.S. compared to the eastern U.S. While regional estimates are of interest, 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 geographic location. Each site is unique. Thus, quantifying the range of background O_3 concentrations at a specific location may be more instructive than determining the range of O_3 concentrations over a large geographic regional scale.

The current analysis as described in the PA (EPA, 2020b, page 2-64) indicates that natural and U.S. anthropogenic O_3 contributions peak during the traditional O_3 season (May through September), while long-range intercontinental transport of international O_3 (i.e., contributions from China, India etc.) peaks in the spring (February through May). The contributions from Canada/Mexico at near-border locations are associated with relatively short-range transport and the seasonality peaks during May through September, like U.S. anthropogenic O_3.

The EPA has indicated in the current PA (EPA, 2020b, pages 2-64 - 2-65) that, based on its latest modeling, the anthropogenic contribution is best correlated with total O_3 at concentrations above 40-50 ppb in both the West and the East, suggesting currently that U.S. anthropogenic emissions are usually the driving cause of high O_3 events at many locations. The EPA does note that there can be exceptions to this rule that are generally associated with natural contributions at high-elevation (e.g., stratospheric-tropospheric transport to the surface), fires events, or at near-border sites. As emissions are reduced, EPA's modeling (EPA, 2014a), as well as empirical data (please see Lefohn et al. (2019b) for specific examples), indicates there is a tendency for the highest hourly average O_3 concentrations to shift toward the spring months (i.e., March-early June). This is the period when the greatest contribution of background O_3 occurs at many locations across the U.S. (Lefohn et al., 2014; EPA, 2014a; Jaffe et al., 2018). The shift toward the spring months appears to indicate that the contribution of background O_3 during the springtime predominates over anthropogenic contributions to ambient O_3 levels.

Figure 1-4. Annual time series of regional average predicted MDA8 total O_3 concentration and contributions of each source (see legend) for the West (top), and the East (bottom). Natural is global natural sources, Intl is international anthropogenic sources, USA is U.S. anthropogenic sources, and Res-Anth is the residual anthropogenic. (Source: EPA, 2020b Fig. 2-23, page 2-49).

In summary, it is important when focusing on margin of safety considerations to understand the range of hourly average background O_3 concentrations. One needs to critically evaluate if human health risk estimates, included in margin of safety considerations, include concentrations within background O_3 levels that play an important role in influencing the total risk estimates. If so, then the risk estimates may be more uncertain than desired to make an adequate margin of safety determination.

1.7 The Dynamics of Changing Hourly Average Concentrations as One Attains the Ozone Standards Focused on the Higher Concentrations

As indicated earlier, as emission reductions occur, the distribution of hourly average concentrations shift from the higher values toward the middle values and the lower values shift upward toward the middle values. There is a compression of the distribution of hourly average O_3 concentrations toward the middle. As a result of the compression, how do we better understand the consequences of how the changes in the distribution of hourly average O_3

concentrations influence the potential effects on human health and vegetation? Understanding the ramifications of the first fundamental principle, which is based on results from controlled human health clinical and vegetation studies, allows us to better place into perspective how the changes in the distribution influence effects estimates. As discussed earlier, the principle is described as follows:

> **Higher Hourly Average O_3 Concentrations Should be Weighted More than Middle and Lower Values when Assessing Human Health and Environmental Effects**.

The Administrator notes (Federal Register, 2020 – page 49842) that in the review completed in 2015 that an 8-hour averaging time remained appropriate for addressing health effects associated with short-term exposures to ambient air O_3 and that it could effectively limit health effects attributable to both short- and long-term O_3 exposures (80 FR 65348, October 26, 2015). Simply stated, by reducing *the higher part of the distribution of hourly average concentrations* (not just the peak hourly values), EPA believed in 2015 that the risk to human health and vegetation would be reduced by reducing the hourly average O_3 concentrations at the upper part of the distribution curve. In addition, the EPA in its 2015 decision (Federal Register, 2015 – pages 65358 – 65359) anticipated that a revised standard with a level of 70 ppb would also reduce the occurrence of exposures to O_3 concentrations at least somewhat below 60 ppb based on its modeling results presented in the 2014 Health Risk and Exposure Assessment document (EPA, 2014b, Figs. 4-9 and 4-10). Thus, even if some members of at-risk populations might experience effects following exposures to O_3 concentrations somewhat below 60 ppb, the Administrator believed in 2015 that a revised level of 70 ppb would be anticipated to reduce the occurrence of such exposures. Thus, in its final 2015 ruling, the EPA considered O_3 exposures that could be relevant for at-risk populations, such as children and people with asthma, and did not agree in 2015 that controlled human exposure studies reporting respiratory effects in healthy adults following exposures to 60 ppb O_3 necessitated a standard level below 70 ppb. 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 revised O_3 standards.

Thus, as emissions were reduced, while the low end of the distribution moved upwards toward the center of the distribution, the EPA believed in 2015 that a revised level of 70 ppb would reduce the occurrence of exposures to O_3 concentrations below 60 ppb and therefore protect human health. In addition, the first key fundamental principle, based on controlled laboratory vegetation experiments published in the 1980s and 1990s, as well as empirical results published in the literature, implies that vegetation effects should not be influenced as the lower concentrations shift upwards toward the mid-level values because of the greater focus on the higher concentrations.

The movement of the lower hourly average O_3 concentrations upward toward the mid-levels as emission reductions occur to attain the O_3 NAAQS helps explain the reasons for the second key fundamental principle. The second principle is described as follows:

Daily Maximum Hourly Averaged O_3 Concentrations Will Remain Well above 0 Parts per Billion (ppb) Even if all Anthropogenic Emissions Were Eliminated Worldwide.

To attain the O_3 standards, a strategy of reducing NO_x emissions has been implemented to reduce the higher hourly average O_3 concentrations. As a result of the reduction of NO titration of O_3 by NO_x reductions, the lower hourly average concentrations at many locations are shifted upward as the more biologically important higher hourly average O_3 concentrations are shifted downward (Simon et al., 2015; Lefohn et al., 2017; Lefohn et al., 2018). 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. As noted in EPA (2020b, page 2-20):

> Simon et al. (2015) found that, similar to results presented in this section for DVs and annual 4th high MDA8 concentrations, the 95th percentile of summertime MDA8 concentrations decreased significantly at most sites across the U.S. between 1998 and 2013. In contrast, trends over that time period for the 5th percentile, median and mean of MDA8 varied with location and time of year. Similarly, Lefohn et al. (2017) reported that between 1980 and 2014 there was a compression of the distribution of measured hourly O_3 values with extremely high and extremely low concentrations becoming less common. As a result, O_3 metrics impacted by high hourly O_3 concentrations, such as the annual 4th highest MDA8 value, decreased at most US sites across this period. Concurrently, metrics that are impacted by averaging longer time periods of hourly O_3 measurements, such as the 6-month (April-September) average of daytime (8am-7pm) O_3 concentrations, were more varied with only about half of the sites exhibiting decreases in this metric and most other sites exhibiting no trend (Lefohn et al., 2017).

In the literature, as well as the EPA's AQS database, there is information that can provide guidance for better understanding the compression of the concentrations discussed above. In many cases for inland monitoring sites, the resulting distribution following emission reductions results in a bell-shaped-like curve. For example, the EPA, in the 1970s, in cooperation with the U.S. Forest Service, established a network of air monitoring stations (referred to as National Air Pollution Background Network (NAPBN)), which was designed to measure levels of O_3 in remote areas within the contiguous 48 states (Evans et al., 1983). There were 8 monitoring sites at various National Forests (NFs) (Green Mt. NF, VT; Kisatchie NF, LA; Custer NF, MT; Chequamegon NF, WI; Mark Twain NF, MO; Croatan NF, NC; Apache NF, AZ; Ochoco NF, OR), which measured O_3, wind speed and direction, temperature, relative humidity, and solar radiation. The network was established to provide a reasonable long-term and continuous record of O_3 concentrations and patterns in areas well removed from anthropogenic sources of air pollution and to make these data available to the EPA and other interested researchers.

Of the 6 NAPBN stations operational for the entire year of 1979, all but the site at Custer NF in Montana recorded hourly average O_3 concentrations which were more than 0.08 ppm. Most days with elevated O_3 concentration, as defined by hourly average values >0.08 ppm, occurred in the spring and early summer months. Evans et al. (1983) hypothesized that the spring events, which occurred at the 7 NAPBN stations, may have been associated with natural sources (i.e., the stratosphere). The Custer NF site was located at Fort Howes in the eastern plains of Montana, near the Wyoming border. Fort Howes is located about 32 km south of Ashland, Montana. Inspecting the hourly average concentration data for the Custer NF site (1250 m, 45° 14' 00" N, 106° 15' 00" W), Fig. 1-5 illustrates that the frequency distribution appears to approach a Gaussian-like (i.e., bell shaped) distribution. Five hourly average concentrations of 75 ppb occurred on 25 April 1979. There were 16 hourly average concentrations at 70 ppb at the site, which occurred on 17 April (4 occurrences), 19 April (2 occurrences), 25 April (5 occurrences), and 26 April (5 occurrences). The two hourly instances on 19 April occurred at 0000 and 0100 in the early morning hours local standard time (LST). The 5 hourly instances of 70 ppb on 26 April occurred during the early morning hours of 0500-0900 LST. In other words, at the Montana site, all the maximum hourly average O_3 concentrations occurred during the springtime and appear to have been related to possible stratospheric-to-tropospheric transport to the surface (STT-S). Without more detailed information concerning the meteorological conditions during the April 1979 periods, it is not possible to definitively associate the highest hourly exposures with natural stratospheric intrusions. However, the occurrence of STT-S processes appear to explain the cause of the elevated hourly O_3 concentrations at the Custer NP site and support the hypothesis stated in Evans et al. (1983).

Lefohn et al. (1998) compared the Custer NF bell-shaped-like frequency distribution (Fig. 1-5) with the distribution of hourly average concentrations for an urban influenced site in Jefferson County, KY (AQS ID 211110027). The frequency distribution of the hourly average O_3 concentrations at the Kentucky site appeared to have a more log-normal-like shape (Fig. 1-6) than the bell-shaped-like distribution experienced at the Custer NF site. In contrast to the Custer NF site, the urban-influenced site in Kentucky showed more high and low hourly average concentrations. Lefohn et al. (1998) noted that the Kentucky site appeared to be influenced by NO titration of O_3 because of the occurrence of more frequent low hourly average concentrations.

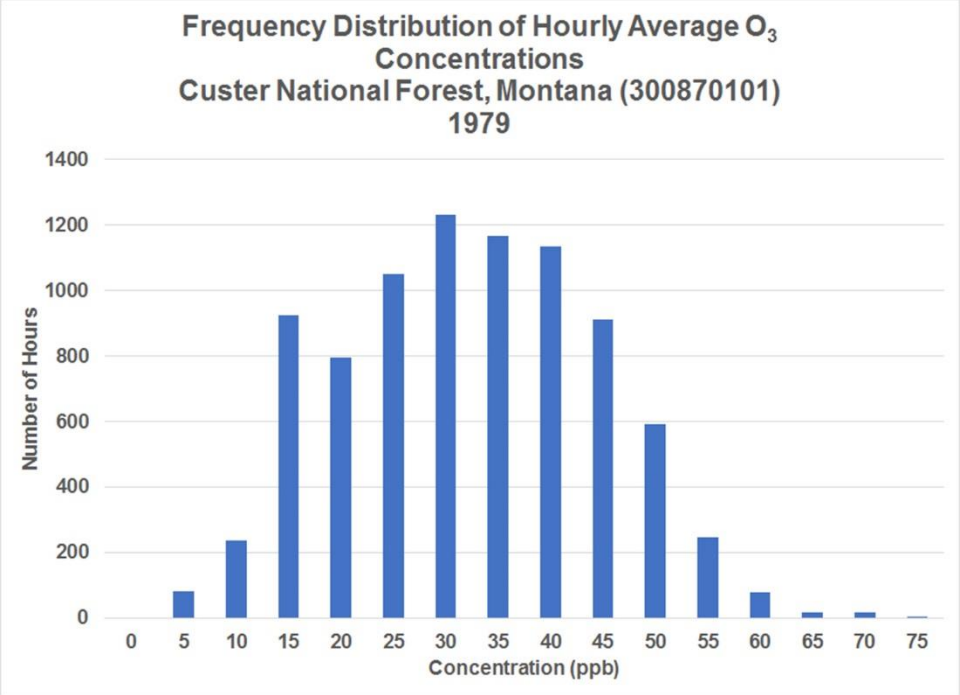


Figure 1-5. Frequency distribution of the hourly average O_3 concentrations in 1979 for Custer National Forest (MT) (300870101) monitoring station. Source of data is from the EPA's AQS database.

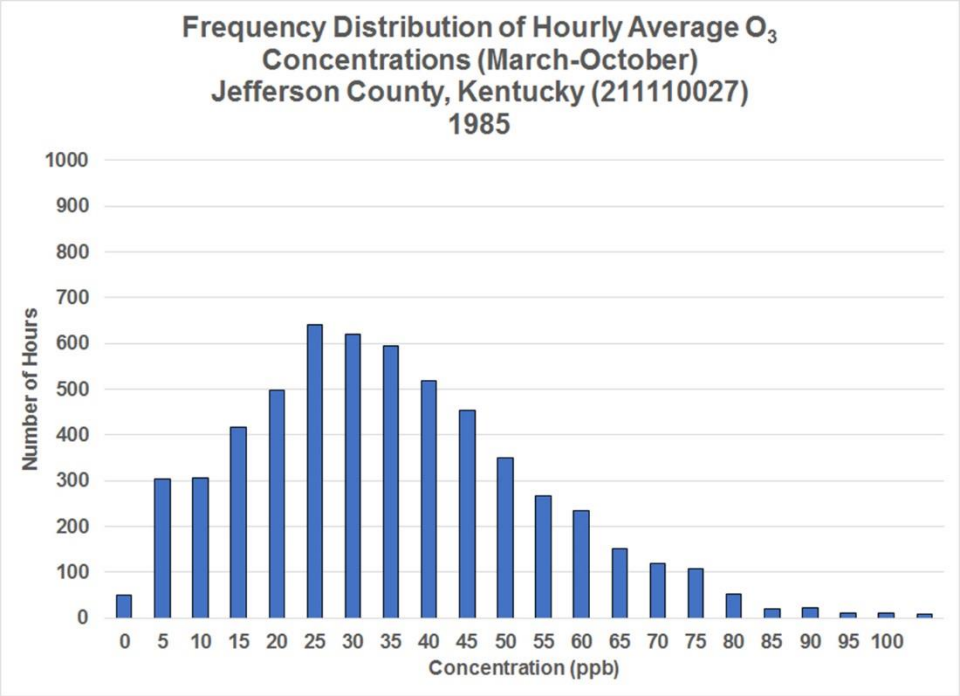


Figure 1-6. Frequency distribution of the hourly average O_3 concentrations in 1985 for an urban-influenced site in Jefferson County (KY) (211110027) monitoring station. Source of data is from the EPA's AQS database.

Based on the comparison of the two sites, Lefohn et al. (1998) hypothesized that it might be possible that as adequate control strategies were implemented to meet the O_3 NAAQS that the distribution pattern of hourly average concentrations for inland monitoring sites might approach the bell-shaped distribution pattern observed at the Montana site, as well as other remote sites in the western U.S. The distribution of hourly average data for the same Kentucky site in 2017 is shown in Fig. 1-7. The distribution shape in 2017 is like the bell-shaped-like distribution observed for the Custer NF site in 1979. Although the site is still influenced by anthropogenic sources, the highest hourly average O_3 concentration has been reduced from 112 ppb (experienced in 1985) to 77 ppb (experienced in 2017). In addition, reviewing the two distribution figures for Jefferson County, Kentucky, shows the compression of the hourly average O_3 concentrations, where the highest hourly average concentrations are moving downward toward the mid-range values and the lowest concentrations are shifting upward toward the mid-range values.

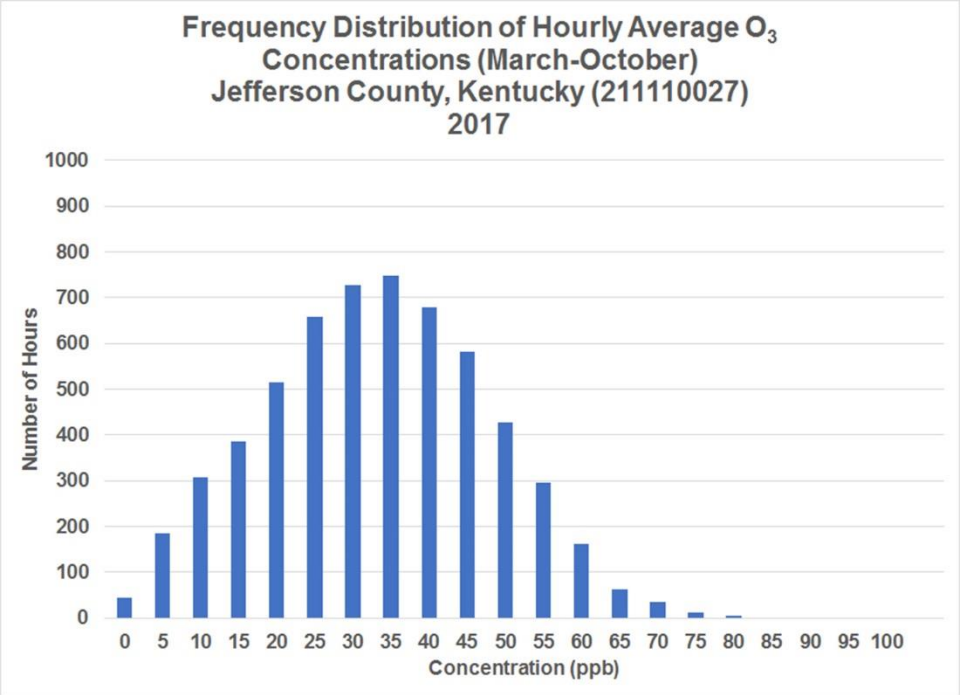


Figure 1-7. Frequency distribution of the hourly average O_3 concentrations in 2017 for an urban-influenced site in Jefferson County (KY) (211110027) monitoring station. Source of data is from the EPA's AQS database.

Thus, these examples illustrate that as emissions are reduced, the distribution of hourly average O_3 concentrations approach a bell-shaped curve, where both the low and high ends of the distribution exhibit minimum frequencies. Additional illustrative examples of the shifting of both the high and low ends of the concentration distribution are provide in Section 3.2.8. The tendency of this phenomenon to occur results in explaining the basis for the second fundamental principle that ***daily maximum hourly averaged ozone concentrations will remain well above 0 parts per billion (ppb) even if all anthropogenic emissions were eliminated worldwide.*** In the next section, we describe important examples of changes in the low and high O_3 concentrations when emissions were reduced during the 2020 COVID-19 lockdowns.

1.8 Recent Empirical Evidence Supporting the Relationship between Reductions of Ozone Precursors and Changes in High and Low Ozone Concentrations – COVID-19 Worldwide Lockdown Emission Reductions

Earlier, it was indicated that as emissions are reduced, the highest hourly average O_3 concentrations are *reduced*, and at many sites the lowest concentrations are *increased* (due to less NO titration of O_3 as NO_x emissions are reduced). Simon et al. (2015) and Sicard et al. (2016) reported more ambient increases in low O_3 concentrations during winter, the time of year where less NO titration of O_3 is most likely to be favored, than during the summer months when O_3 production efficiency is high (Lefohn et al., 2017). This phenomenon has been observed at many locations across the U.S. (Simon et al., 2015; Lefohn et al., 2017, 2018; EPA, 2020b). The same phenomenon has also been observed at sites outside of the U.S. (Lefohn et al., 2018).

Because of the unfortunate COVID-19 pandemic, many countries around the world during the spring (northern hemisphere) and fall (southern hemisphere) of 2020 instituted immediate lockdown orders. As a result of these orders, anthropogenic emissions were severely reduced. Recognizing the opportunity to investigate how severe emission reductions influenced air pollution concentrations, researchers documented these changes. One important result of the observations documented over the short period in 2020, as emissions were reduced, was additional confirmation about the changes that occurred in the high concentration part of the distribution, as well as the low part of the distribution. These observations are important because they provide insight concerning changes in O_3 distribution patterns as emissions are reduced to attain current and future NAAQS standards to protect human health and welfare.

In earlier sections, the two fundamental principles were described. The first fundamental principle (i.e., **Higher Hourly Average O_3 Concentrations Should be Weighted More than Middle and Lower Values when Assessing Human Health and Environmental Effects**) focuses on the higher hourly average concentrations within the distribution. The second fundamental principle (i.e., **Daily Maximum Hourly Averaged O_3 Concentrations Will Remain Well above 0 Parts per Billion (ppb) Even if all Anthropogenic Emissions Were Eliminated Worldwide**) focuses on the changes in the entire distribution of hourly average concentrations as emissions are reduced. During the 2020 COVID-19 lockdown, scientists characterized changes in the O_3 levels using combinations of exposure metrics. Some of the investigators used metrics that focused on the lower end of the distribution, while other researchers applied metrics that focused on the upper end. In some cases, the investigators reported *increasing* O_3 during lockdown, while other researchers reported *decreasing* O_3. As will be discussed below, there are possible reasons responsible for explaining the increases and decreases in O_3 concentrations during lockdown.

Sicard et al. (2020) reported on the effect of lockdown due to the COVID-19 pandemic on air pollution in four Southern European cities (Nice, Rome, Valencia, and Turin) and Wuhan, China. The focus of their study was on O_3. Compared to the same period in 2017-2019, the daily O_3 mean concentrations increased at urban stations by 24% in Nice, 14% in Rome, 27% in Turin, 2.4% in Valencia, and 36% in Wuhan during the lockdown in 2020. According to Sicard et al.

(2020), the increase in O_3 concentrations was mainly explained by an unprecedented reduction in NO_x emissions leading to a lower O_3 titration by NO.

Huang et al. (2020) reported the results of imposed nationwide lockdown restrictions in China after the Chinese New Year in January of 2020. The authors reported that despite large decreases in primary pollution, there were several periods of heavy haze pollution in East China during the COVID-19 lockdown, raising questions about the well-established relationship between human activities and air quality. The authors calculated differences in averaged concentrations of NO_2, $PM_{2.5,}$ O_3, and the $PM_{2.5}$/CO ratio prior to lockdown (January 2 – January 23, 2020) and during lockdown (January 26 – February 17, 2020). Huang et al. (2020) reported that large decreases in NO_x emissions occurred during lockdown from transportation with increases in average O_3 concentrations and nighttime NO_3 radical formation, and these increases in atmospheric oxidizing capacity in turn facilitated the formation of secondary inorganic and organic particulate matter.

Le et al. (2020) reported for eastern China that up to 90% reduction of certain emissions during the city-lockdown period were identified from satellite and ground-based observations. The primary focus period during the COVID-19 lockdown in China was from January 23 to February 13, 2020. This period encompassed a 7-day national holiday traditionally celebrating the Lunar New Year, during which previous studies have noted the reduction in anthropogenic emissions. Unexpectedly, extreme particulate matter levels simultaneously occurred in northern China. The author's synergistic observation analyses and model simulations showed that anomalously high humidity promoted aerosol heterogeneous chemistry, along with stagnant airflow and uninterrupted emissions from power plants and petrochemical facilities, contributing to severe haze formation. In addition, Le et al. (2020) noted that because of non-linear production chemistry and titration of O_3 in winter, reduced nitrogen oxides resulted in O_3 enhancement of 3-week average concentrations in urban areas, further increasing the atmospheric oxidizing capacity and facilitating secondary aerosol formation.

Mahato et al. (2020), described the results of a nationwide lockdown in Delhi, India initially from March 24 to April 14, 2020 and extended up to May 3, 2020. With the aid of air quality data of seven pollutant parameters (PM_{10}, $PM_{2.5}$, SO_2, NO_2, CO, O_3, and NH_3) for 34 monitoring stations spread over the megacity, the authors evaluated the spatial pattern of air quality in pre and during-lockdown phases. The results demonstrated that during lockdown air quality was significantly improved. Among the selected pollutants, concentrations of PM_{10} and $PM_{2.5}$ were reduced (>50%) in comparison to the pre-lockdown phase. In comparison to the previous year (i.e., 2019) during the same time the reduction of PM_{10} and $PM_{2.5}$ was as high as about 60% and 39%, respectively. Among other pollutants, NO_2 (−52.68%) and CO (−30.35%) level were also reduced during the lockdown phase. About 40% to 50% improvement in air quality was identified just after four days of commencing lockdown. Using the average of the daily 8-h average concentration across sites, the authors noted that the concentration of O_3 increased in the industrial and transport dominated locations (>10% increase). Mahato et al. (2020) noted that the cause for the increase in O_3 concentration, especially in the industrial and transport dominated areas, was the decrease of NO, which led to the lowering of the O_3 consumption (titration, $NO + O_3 = NO_2 + O_2$) and caused an increase in O_3 concentrations.

Dantas et al. (2020) described the impact of COVID-19 partial lockdown during the fall period on the air quality of the city of Rio de Janeiro, Brazil. On March 16, 2020, the state's governor declared a public health emergency in the city of Rio de Janeiro and partial lockdown measures came into force a week later. The pre-lockdown period was March 2 – 22, 2020 and lockdown was from March 23 – April 16. The authors compared the particulate matter, carbon monoxide, nitrogen dioxide, and O_3 concentrations determined during the partial lockdown with values obtained in the same period of 2019 and with the weeks prior to the virus outbreak. For the comparison of the results obtained in different days, median concentrations of hourly average values were used in their analyses. Concentrations varied with substantial differences among pollutants and among the three studied monitoring stations. According to the authors, CO levels showed the most significant reductions (30.3–48.5%) since they were related to light-duty vehicular emissions. The authors noted that NO_2 showed reductions, while PM_{10} levels were only reduced during the first lockdown week. Dantas et al. (2020) reported that in April, an increase in vehicular flux and movement of people was observed mainly because of the lack of consensus about the importance and need of social distancing and lockdown. The authors noted that O_3 concentrations increased probably due to the decrease in the nitrogen oxides level. When compared to the same period of 2019, NO_2 and CO median values were 24.1–32.9 and 37.0–43.6% lower. The authors cautioned that meteorological interferences, mainly the transport of pollutants from the industrial areas, might have influenced the results.

Patel et al. (2020) presented a case study from Auckland, New Zealand, an isolated southern hemisphere city, which is largely unaffected by long-range pollution transport or industrial sources of air pollution. In this city, traffic flows reduced by 60–80% because of a government-led initiative to contain the virus by limiting all transport to only essential services. The authors characterized changes in ambient pollutant concentrations of NO_2, O_3, BC, $PM_{2.5}$, and PM_{10} between the lockdown period (March 27 – April 17, 2020), with data from this period compared with data for a similar time of year (February to April) extending back to 2015 for assessing consideration of the impacts of the local meteorology on air pollution levels at the same time of year. Using 24-hour average air pollution concentrations, statistically significant changes were observed before and after lockdown at two sites for all pollutants evaluated, based on t-tests. Patel et al. (2020) showed that the source emission reductions had significant but non-linear impacts on air quality. While emission inventories and receptor modelling approaches confirm the dominance of traffic sources for NO_x (86%), and BC (72%) across the city, observations suggest a consequent reduction in NO_2 of only 34–57% and a reduction in BC of 55–75%. While these two pollutants were reduced, O_3 concentrations increased. The authors noted that a lesser increase in O_3 would be anticipated due to the already low background levels of O_3 observed in the southern hemisphere. The observed reductions in $PM_{2.5}$ (still likely to be dominated by traffic emissions), and PM_{10} (dominated by sea salt, traffic emissions to a lesser extent, and affected by seasonality) were found to be significantly less (8–17% for PM2.5 and 7–20% for PM_{10}).

In the U.S. during the lockdown period, substantial anthropogenic emissions were reduced. To better understand the spatial extent of reductions of nitrogen oxides across the U.S., an overview summary figure is presented. Goldberg et al. (2020) used TROPOMI satellite data to illustrate substantial drops in NO_2 during COVID-19 physical distancing between 2020 and 2019. The authors used three different methods. In Method 1, they compared an average of 15

March – 30 April 2020 to the same timeframe of 2019 and account for impact of changes due to solar zenith angle. In Method 2, the authors develop a strategy to account for varying weather patterns without the use of a chemical transport model. In this method, they normalize each day's NO_2 observation to a day with "standard" meteorology—like standard temperature and pressure (STP) conditions in a laboratory setting. They accounted for four different day-varying effects (solar zenith angle, wind speed, wind direction, and day-of-week). In Method 3, the authors inferred a TROPOMI NO_2 column amount assuming no COVID-19 precautions using the GEM-MACH regional chemical transport model, which was operationally run in forecast mode. The authors then compared the actual TROPOMI columns to the theoretical columns. Methods 2 and 3 both accounted for year-varying meteorology, while Method 1 did not.

The authors reported that meteorological patterns were especially favorable for low NO_2 in much of the U.S. during spring 2020, complicating comparisons with spring 2019. After accounting for solar angle and meteorological considerations, the authors calculated that NO_2 declines ranged between 9.2% and 43.4% among 20 cities in North America, with a median of 21.6%. Of the studied cities, largest NO_2 drops (>30%) were in San Jose, Los Angeles, and Toronto, and smallest declines (<12%) were in Miami, Minneapolis, and Dallas. The authors pointed out that normalized NO_2 changes could be used to highlight locations with greater activity changes and better understand the sources contributing to adverse air quality in each city. For illustrative purposes, Fig. 1-8 is presented using Method 1. Note that the largest decreases in NO_2 were near major cities in North America. The authors reported regional decreases in eastern North America. On the other hand, Central and Northwestern U.S. appeared to have seen little change between years. If one wishes to assess the potential for long-range transport to the U.S., it is important to note that the lockdown period for China was approximately from January 23 to February 13, 2020 (Le et al., 2020). The lockdown in India was imposed initially for three weeks from March 24 to April 14, 2020 and extended to May 3, 2020 (Mahato et al. (2020). Goldberg et al. (2020) defined the "post-COVID-19" period as the timeframe when COVID-19 precautions were instituted, which was defined to be initiated on March 15. According to the authors, the period was plus or minus a few days in various U.S. cities. Figure 1-8 illustrates the column NO_2; in rural areas, column NO_2 changes may be de-coupled to near-surface NO_2 changes.

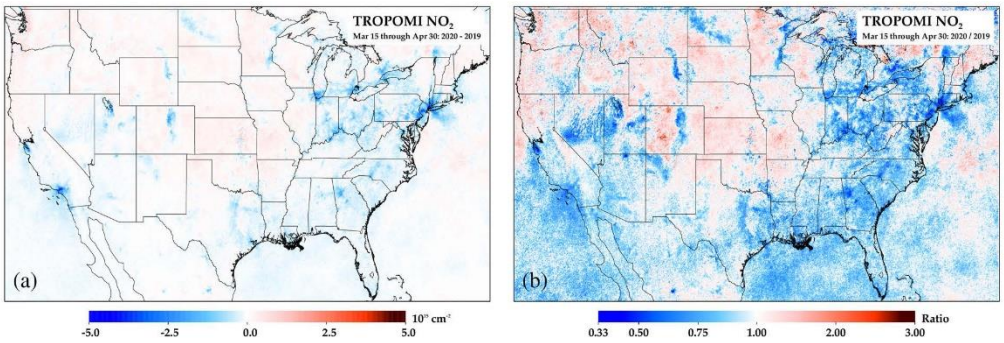


Figure 2. TROPOMI NO_2 differences between 2019 and 2020, using 15 March to 30 April 2020 as the post-COVID-19 period. Plots are showing (a) the absolute difference and (b) the ratio between years.

Figure 1-8. TROPOMI NO_2 differences between 2019 and 2020, using the 15 March to 30 April 2020 as the post-COVID-19 period using Method 1 as defined in the study. Plots are showing (a) the absolute difference and (b) the ratio between years. Source: Goldberg et al. (2020).

Chen et al. (2020) described the impacts of COVID-19 lockdown on air quality over the U.S. Many state governments, but not all, in the U.S. issued lockdown or business restrictions amid the COVID-19 pandemic in March 2020. Using air quality data for 28 sites, their analyses revealed widespread but nonuniform reductions of NO_2 and CO during the first phase of lockdown (March 15–April 25, 2020) relative to a pre-lockdown baseline periods (January 25 – March 7, 2020, as well as the same period for 2017–2019). The authors applied a ratio of the mean concentrations at each site during two time periods (P1 defined over the period March 15 – April 25 and P0 defined for the period January 25 – March 7, 2020). The ratio was then compared to similar ratios derived for the baseline period for 2017, 2018, and 2019. The authors reported that the reductions, up to 49% for NO_2 and 37% for CO, were statistically significant at two thirds of the sites and tended to increase with local population density. According to the authors, significant reductions of particulate matter ($PM_{2.5}$ and PM_{10}) only occurred in the Northeast and California/Nevada metropolises, where NO_2 declined the most, while the changes in O_3 concentrations were mixed and relatively minor.

Using data from the EPA databases, Sommer et al. (2020) reported O_3 decreases at many U.S. locations. Sommer et al. (2020), in a National Public Radio (NPR) article, noted that O_3 concentrations decreased during the spring lockdown of 2020 at many locations compared to spring levels for the years 2015, 2016, 2017, 2018, and 2019. The authors compared the median value for the daily maximum 8-h average concentrations detected during March 15 – April 30, 2020 with levels reported during the comparable period over the previous five years (2015-2019). The authors pooled all the data for the qualifying monitors within a given CBSA together and compared the 2020 median values with the previous 5 years. Their analysis revealed that, in most locations, the median O_3 concentrations decreased by 15% or less. Figure 1-9 below, summarizes the results of their analysis. For the areas investigated, O_3 reductions occurred because of lockdowns in parts of the West, Midwest, and East. According to the authors, during the lockdown period, a series of rainstorms swept through southern California in March, which helped remove pollutants from the air. The Seattle area showed modest declines in O_3 levels,

while the Portland area experienced small reductions in O_3 concentrations. The Northeastern U.S. experienced little change. The Mid-Atlantic states experienced modest declines in O_3 levels. In comparing Fig. 1-8, illustrating that the NO_2 changes from Goldberg et al. (2020), with Fig. 1-9, it appears that the spatial patterns for NO_2 reductions were similar to the spatial patterns of change observed for O_3 by Sommer et al. (2020).

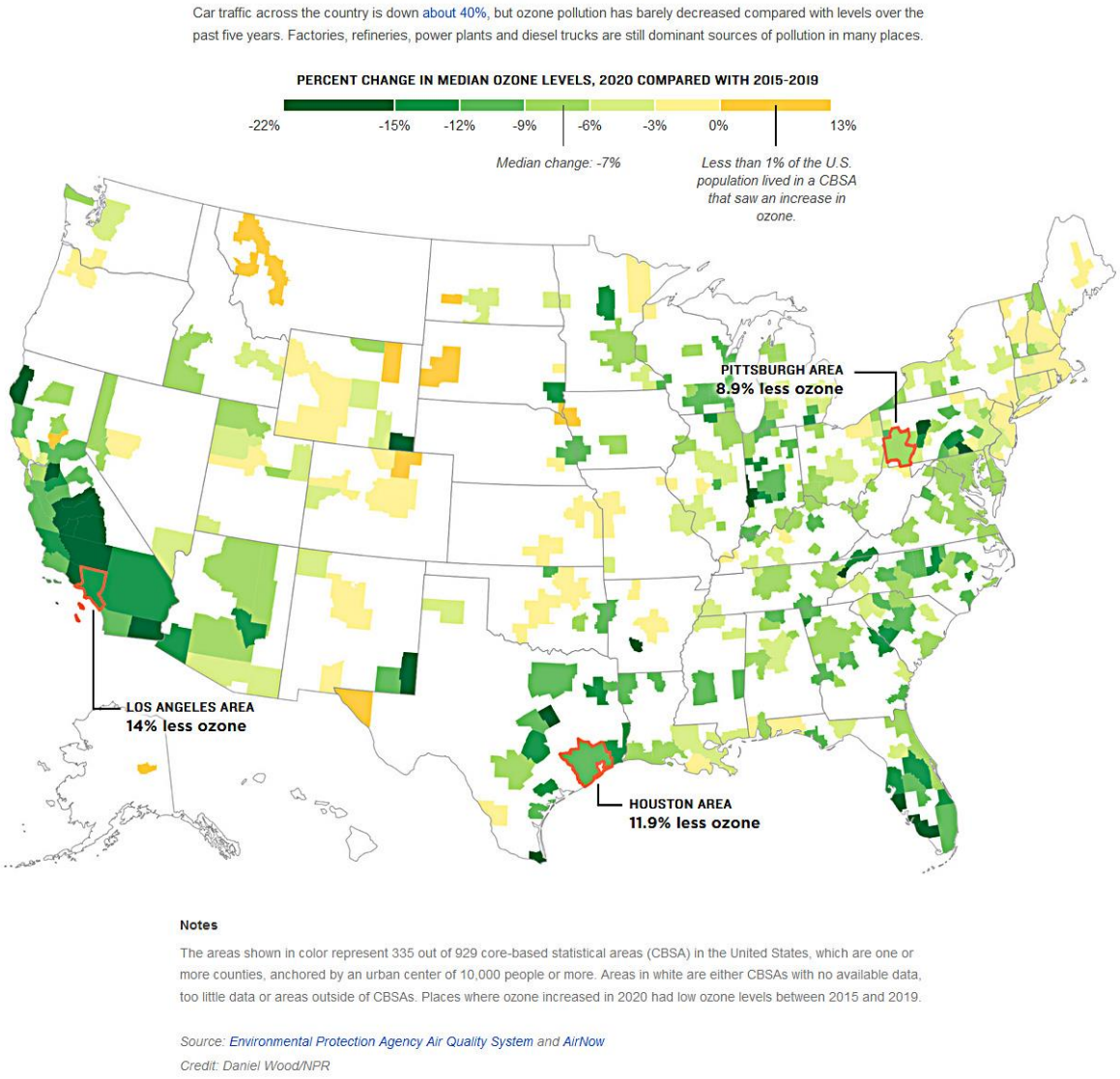


Figure 1-9. Percent change in median ozone levels of daily maximum 8-h average concentrations, with 2020 spring levels compared with 2015-2019. Source: Sommers et al. (2020). National Public Radio article (May 19, 2020). Traffic Is Way Down Because of Lockdown, But Air Pollution? Not So Much. https://www.npr.org/sections/health-shots/2020/05/19/854760999/traffic-is-way-down-due-to-lockdowns-but-air-pollution-not-so-much.

Using the same O_3 metric (the median value of the daily maximum 8-h average concentrations) applied by Sommer et al. (2020), daily maximum 8-h average concentrations from the EPA AQS and AirNow databases were downloaded for 52 U.S. O_3 monitoring sites for the period March 15 – April 25, 2020. For most of the 52 monitoring sites, the EPA-defined O_3 season contained the months of March and April. The median values characterized by site for the March and April period in 2020 were compared with the same period for 2017, 2018, and 2019. Changes between 5% and -5% were subjectively defined as NO CHANGE because of possible year-to-year variability of meteorological factors, as well as year-to-year variability of stratospheric-tropospheric transport to the surface (STT-S). For this analysis, it was not possible to take into consideration these, as well as other, variables. Table 1-1 summarizes for 28 O_3 monitoring sites in the West the changes that occurred during lockdown.

Table 1-1. Percent change in median ozone levels of daily maximum 8-h average concentrations in the West for 28 monitoring sites, with 2020 March 15 – April 25 levels compared with 2017-2019.

City	State	AQS ID	Percent Change	Direction of Change
Denali NP	AK	020680003	-4	NO CHANGE
Seattle	WA	530330080	-6	DECLINE
Mount Rainier NP	WA	530530012	-7	DECLINE
Portland	OR	410510080	-2	NO CHANGE
Los Angeles	CA	060371103	-21	DECLINE
Joshua Tree NP	CA	060719002	-12	DECLINE
San Jose	CA	060850005	-7	DECLINE
Fresno	CA	060190011	-23	DECLINE
Sequoia & Kings Cany NP	CA	061070009	-16	DECLINE
Lassen Volcanic NP	CA	060893003	-10	DECLINE
Redding	CA	060890004	-6	DECLINE
Las Vegas	NV	320030540	-8	DECLINE
Great Basin NP	NV	320330101	0	NO CHANGE
Phoenix	AZ	040139997	-16	DECLINE
Chiricahua NM	AZ	040038001	-7	DECLINE
Grand Canyon NP	AZ	040058001	-10	DECLINE
Albuquerque	NM	350010023	-9	DECLINE
Glacier NP	MT	300298001	-5	NO CHANGE
Helena	MT	300490004	-1	NO CHANGE
Cheyenne	WY	560210100	-19	DECLINE
Yellowstone NP	WY	560391011	-3	NO CHANGE
Denver	CO	080590006	-2	NO CHANGE
Denver	CO	080350004	-3	NO CHANGE
Mesa Verde NP	CO	080830101	-6	DECLINE
Rocky Mountain NP	CO	080690007	-4	NO CHANGE
Salt Lake City	UT	490353006	-3	NO CHANGE
Canyonlands NP	UT	490370101	-8	DECLINE
Dinosaur NM	UT	490471002	-5	NO CHANGE

Sites, such as Denali National Park (AK), Glacier National Park (MT), and Yellowstone National Park (WY), which experience little locally generated pollution, experienced no change in O_3 concentrations during lockdown. Similar to the results reported by Sommer et al. (2020), the Seattle area showed modest declines in O_3 levels, while the Portland area experienced small

reductions in O_3 concentrations. Table 1-2 summarizes for 10 O_3 monitoring sites in the Midwest, the changes that occurred during lockdown when compared to the same period for 2017 – 2019. Similar patterns of change occurred in the Midwest in this analysis compared to the Sommer et al. (2020) results. Table 1-3 summarizes for 14 O_3 monitoring sites in the East the changes that occurred during lockdown when compared to the same period for 2017 – 2019. The Mid-Atlantic states experienced modest declines in O_3 levels. This analysis showed little changes in O_3 concentrations occurring in the Northeastern U.S., which was similar to those noted by Sommer et al. (2020).

Table 1-2. Percent change in median ozone levels of daily maximum 8-h average concentrations in the Midwest for 10 monitoring sites, with 2020 March 15 – April 25 levels compared with 2017-2019.

City	State	AQS ID	Percent Change	Direction of Change
Bismarck	ND	380150003	-11	DECLINE
Minneapolis	MN	270031002	-9	DECLINE
Chicago	IL	170310001	-9	DECLINE
Indianapolis	IN	180970078	-10	DECLINE
Kansas City	KS	202090021	-1	NO CHANGE
St. Louis	MO	295100085	-9	DECLINE
Tulsa	OK	401431127	-12	DECLINE
Houston	TX	482010024	-23	DECLINE
Big Bend NP	TX	480430101	-12	DECLINE
Chamizal Nat. Memorial.	TX	481410044	1	NO CHANGE

Table 1-3. Percent change in median ozone levels of daily maximum 8-h average concentrations in the East for 14 monitoring sites, with 2020 March 15 – April 25 levels compared with 2017-2019.

City	State	AQS ID	Percent Change	Direction of Change
Cincinnati	OH	390610040	-12	DECLINE
Cleveland	OH	390350060	-11	DECLINE
Pittsburgh	PA	420030067	-6	DECLINE
Boston	MA	250250042	-3	NO CHANGE
Cape Cod Nat. Seashore	MA	250010002	-2	NO CHANGE
Providence	RI	440071010	-7	DECLINE
New York	NY	360810124	0	NO CHANGE
Bellevue State Park	DE	100031013	-6	DECLINE
Grantville	MD	240230002	-13	DECLINE
District of Columbia		110010043	-2	NO CHANGE
Richmond	VA	510870014	-6	DECLINE
Raleigh	NC	371830014	-12	DECLINE
Great Smoky Mountain NP	TN	470090101	-7	DECLINE
Atlanta	GA	130890002	-13	DECLINE

As noted above, the cause for the increase in O_3 concentration at some locations during lockdown, especially in the industrial and transport dominated areas, was associated with the decrease of NO, which led to the lowering of the O_3 consumption (titration, $NO + O_3 = NO_2 + O_2$). As emissions are reduced, Simon et al. (2015) note that in the U.S., increasing O_3 trends generally occur in the winter months (defined by the authors as October-April) in more urbanized areas and at the lower end of the O_3 distribution. As indicated in this section, the response of O_3 concentrations to lockdown varied at many locations around the world. Some of the variation could be associated with the selection of the specific exposure metric used to investigate changes in O_3 concentrations. Some of the investigators selected 24-h average concentrations, while others selected longer-term averaged hourly concentrations, long-term average of daily maximum 8-h average O_3 concentrations, median concentration hourly average values, the ratio of mean concentrations, and the median O_3 levels of the daily maximum 8-h average concentrations. Some of these metrics focus on the lower O_3 concentrations, while other metrics focus on the higher O_3 concentrations. Using the same hourly average O_3 concentration data, Lefohn et al. (2017, 2018) have discussed how the selection of different exposure metrics result in different trend outcomes.

As discussed earlier, as emission reductions occur, the different parts of the distribution of hourly average O_3 concentrations shift differently. Both ends of the distribution of hourly average O_3 concentrations shift toward the middle of the distribution. The lower concentrations shift upward, while the higher concentrations shift downward. Based on the metric selected to characterize changes in O_3 exposures during lockdown compared to previous years, different results would be anticipated. A good question to ask is: "Which O_3 exposure metric is most appropriate when comparing differences between exposures during lockdown and other periods?" The answer is that it depends upon that the question being asked. For example, if one were interested in exploring the answer to the question: "What is the benefit to public health by lowering emissions associated with O_3 concentrations," one would focus on a metric associated with the higher part of the O_3 distribution of hourly average concentrations. This is because as noted in earlier discussions, results from controlled human health exposure studies showed that the higher hourly average O_3 concentrations were associated greater effects. Alternatively, if one were interested in the chemical phenomena associated with how O_3 concentrations change as NO_x concentrations were reduced during lockdown, one might ask the question: "Do the lower hourly average O_3 concentrations increase or decrease due to NO_x reductions?" The first question deals with public health, while the second focuses on how chemical changes in the atmosphere influence the changes in the lower hourly average concentrations. Thus, if one were interested in how emission reductions during lockdown affected public health, one should focus on O_3 exposure indices that highlight changes in the higher concentrations. One such index is the daily maximum of the 8-h average concentration. For health and vegetation concerns, focus should be on the change in the frequency of the upper part of the distribution of concentrations at specific monitoring sites.

The "natural experiment", a term used by Patel et al. (2020), showed that large source emission reductions had significant impacts on air quality across the world. The reported worldwide results illustrated that during lockdown as emissions were reduced the lower concentrations shifted upward. The shifting of the lower concentrations upward as extreme emission reductions occurred during lockdown provides additional evidence that support the key

fundamental principle 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**.

1.9 The adequacy of the 4th highest daily 8-h maximum concentration exposure metric as a substitute for the W126 exposure index to protect vegetation

1.9.1 Introduction

The EPA (EPA 2020b, page 4-34) focused its analyses in the last several reviews (Federal Register, 2007, page 37818; Federal Register, 2008, page 16436; Federal Register, 2015, pages 65373-65374) on vegetation exposure metrics that characterize cumulative exposures over a season or seasons (i.e., 3 years). The use of the W126 to characterize O_3 exposure concentrations regarding potential vegetation effects, particularly growth, has received strong support from CASAC in previous reviews (Henderson, 2006; Samet, 2010; Frey, 2014).

In August 2019, the U.S. Court of Appeals for the District of Columbia Circuit (DC Circuit) addressed arguments regarding the adequacy of the EPA Administrator's 2015 decision to use a 3-year average of the W126 index as the benchmark and use the current form of the standard instead of the CASAC recommended W126 metric. The Court noted:

> The Environmental Petitioners raise two challenges pertaining to the W126 index, both as a benchmark and as a form and averaging time. First, they argue that EPA impermissibly departed from CASAC's advice by setting the secondary standard level using a three-year average W126 benchmark without lowering the level to protect against single-year exposures associated with median annual tree growth loss of 6%, which CASAC had advised was "unacceptably high." J.A. 518. They also contend that EPA arbitrarily disregarded CASAC's advice to adopt the W126 index as the form and averaging time for the secondary standard. We remand to EPA on the first issue and do not reach the second.
>
> The Three-Year Average Benchmark. CASAC advised EPA that basing the secondary standard on a "single-year period" would provide "more protection for annual crops and for the anticipated cumulative effects on perennial species" than a three-year average. J.A. 518. It explained that EPA's proposal to use a three-year averaging period was "not supported by the available data," J.A. 536, and that if EPA chose to "base the secondary standard on a three-year averaging period," then "the level of the standard should be revised downward such that the level for the highest three-month summation in any given year of the three-year period would not exceed [its] scientifically recommended" range of single-year, W126 exposure levels, J.A. 518. This was necessary, CASAC explained, to "protect against single unusually damaging years that will be obscured in the average." J.A. 536.

EPA argues it gave effect to CASAC's recommendation by using a three-year average benchmark of 17 ppm-hrs, which, after adjusting for the cottonwood data, was "somewhat below" the 19 ppm-hrs associated with 6% annual growth loss that CASAC had advised was "unacceptably high." 80 Fed. Reg. at 65,406-07 (quoting J.A. 518). But CASAC had advised a maximum level associated with 5.2% annual biomass loss, see J.A. 631, and it expressly cautioned that 6% median growth loss in a single year was unacceptable, see J.A. 518. EPA's use of a benchmark that averages out to less than 6% biomass loss over three years does not accord with CASAC's advice. Indeed, as commenters informed EPA during the rulemaking, see J.A. 1836-40, EPA's own air quality data suggests that many large national parks and wilderness areas that have met EPA's chosen three-year average 17 ppm-hrs benchmark— areas that Congress considers significant to the public welfare, see 42 U.S.C. §§ 7470(2), 7472(a)—have meanwhile recorded single-year W126 values at and above 19 ppm-hrs, which is associated with "unacceptably high" annual biomass loss of 6% and higher. See J.A. 1061-64; 80 Fed. Reg. at 65,391. EPA critiques that data as marred by outdated handling procedures, but the agency acknowledged that other data derived through updated procedures produced results "similar to" those showing harmful exposure spikes. J.A. 1213. Critically, EPA points us to no data or analysis (based on new or old procedures) suggesting that the chosen benchmark prevents single seasonal exposures of 19 ppm-hrs or higher. In short, EPA has not demonstrated how its chosen benchmark protects against "unusually damaging years that will be obscured in the average." J.A. 536.

EPA alternatively defends its decision to use the three-year-average benchmark as providing a focus on public welfare effects of "potentially greater" significance than effects "associated with a single year" of exposure. 80 Fed. Reg. at 65,404. This position, however, is inconsistent with EPA's other actions. In establishing the secondary standard, for example, EPA heavily relied on data showing annual 6% median tree growth loss at 19 ppm-hrs, see id. at 65,406, and acknowledged the potential for a single season of high ozone exposure to "alter biomass allocation and plant reproduction in seasons subsequent to [that season's] exposure," thereby leading to "a negative impact on species regeneration in subsequent years," id. at 65,371-72; see also J.A. 740-41. EPA additionally recognized that "ozone effects in plants are cumulative," id. at 65,373 (quoting EPA, Integrated Science Assessment 2-44 (2013)), meaning that the adverse vegetative effects from single, high-ozone years are not offset by subsequent low-ozone years. EPA has identified no contrary evidence in the record demonstrating why these single-year effects matter less than a three-year average.

We therefore remand this issue for EPA to either lower the standard to protect against unusually damaging cumulative seasonal exposures that will be obscured in its three-year average, or explain its conclusion that the unadjusted average is an appropriate benchmark notwithstanding CASAC's contrary advice. Alternatively, EPA could adopt the single-year W126 exposure index as the form

> and averaging time, which would presumably moot any problems with the way it translated that index to use as a benchmark.
>
> The Form and Averaging Time. CASAC also recommended that EPA use the single-year W126 index as the form and averaging time for the secondary standard. J.A. 518. EPA instead chose to retain the existing form and averaging time—the three-year average of the fourth-highest daily maximum eight-hour concentration. Adopting the W126 index as the form and averaging time was unnecessary, EPA explained, because the ozone exposure levels associated with the existing form and averaging time and a three-year average of the W126 index are "highly correlated," especially at lower levels, and "future control programs designed to help meet a primary [ozone] standard based on the" current form and averaging time should "provide similar improvements in terms of the 3-year average of the annual W126 metric." J.A. 1253; see also 80 Fed Reg. at 65,400-01, 408-09.
>
> The Environmental Petitioners argue that EPA did not justify its decision not to adopt the W126 index as the form and averaging time. We lack any basis to assess the reasonableness of EPA's actions, however, because a critical piece of the puzzle is missing. To review: EPA chose not to use the W126 index as the form and averaging time because it found that ozone exposure levels associated with the existing form and averaging time are "highly correlated" to a three-year average of the W126 index. But, as discussed, EPA never explained why it is reasonable to focus on a three-year average of the W126 index in the first place. Therefore, we cannot assess the relevance of the claim that the two are "highly correlated." EPA's reconsideration on remand of the three-year averaging issue should supply us with the information necessary to resolve this question, or, if EPA chooses to follow CASAC's advice to lower the standard to control for unusually high single years, potentially moot the Environmental Petitioners' concern that the current form tolerates even three-year average W126 levels higher than 17 ppm-hrs during periods when a 0.07 ppm, 8-hour level is met. Accordingly, we decline to reach this question.

The CASAC was clear on its recommendations to the Administrator concerning the use of the W126 exposure index as a standard to protect vegetation. In its 2014 letter to the Administrator (Frey, 2014) concerning the draft PA (EPA, 2014) regarding the substitution of the W126 exposure index with the 8-h primary standard, CASAC indicated within its letter:

> The CASAC concurs with the justification in this section that the form of the standard should be changed from the current 8-hr form to the cumulative W126 index and finds that the discussion provides an appropriate and sufficient rationale.
>
> This section clearly demonstrates that ozone-induced injury may occur in areas that meet the current standard. As noted above, the correlative similarity between the current standard and a level of the W126 index of 15 ppm-hrs must not be

interpreted to mean that just meeting the current standard is equivalent to just meeting a W126 level of 15 ppm-hrs. Most of the analyses found effects below 15 ppm-hrs (many at 10 or even 7 ppm-hrs). Based on review of relevant science, the CASAC concludes that the upper bound of the range that should be considered for the W126 standard should not exceed 15 ppm-hrs. The CASAC does not support a level higher than 15 ppm-hrs. Levels above 15 ppm-hrs should not be included in the revised PA as options for an alternate secondary standard. For example, at 17 ppm-hrs, the median tree species has 6% relative biomass loss, and the median crop species has over 5% loss. These levels are unacceptably high.

The CASAC does not recommend the use of a three-year averaging period. We favor a single-year averaging period, which will provide more protection for annual crops and for the anticipated cumulative effects on perennial species. The scientific analyses considered in this review, and the evidence upon which they are based, are from single-year results. If a 3-year averaging period is established, then the upper limit will need to be reduced to protect against one-year ozone peaks. We consider this further in the response to charge questions for Chapter 6.

The suggestion in Section 6.2 to use a 3-year averaging period is not supported by the available data. We have not supported it in the past nor do we support it here. The primary justification for a 3-year averaging period is to improve the program stability of the classification of regions as being in or out of compliance. The proposed form includes a 3-month period, so it is not nearly as sensitive to extreme events as an hourly or 8-hour averaging period. The case has not been made that welfare benefits from the stability of a 3-year average are greater than those from using the biologically relevant 1-year value. If a 3-year averaging period is implemented, it should be at a lower level than a single-year standard to protect against single unusually damaging years that will be obscured in the average.

In reaching its scientific judgment regarding the indicator, form, averaging time, and range of levels for a revised secondary standard, the CASAC has focused on the scientific evidence for the identification of the kind and extent of adverse effects on public welfare. The CASAC acknowledges that the choice of a level within the range recommended based on scientific evidence is a policy judgment under the statutory mandate of the Clean Air Act. Specifically, the Clean Air Act grants discretion to the Administrator to specify a standard that is “requisite to protect the public welfare from any known or anticipated adverse effects associated with the presence of [the] pollutant in the ambient air” (Section 302(h), 42 U.S.C., §7602(h)). As a policy recommendation, separate from its advice above regarding scientific findings, the CASAC advises that a level of 15 ppm-hrs is requisite to protect crop yield loss, but that lower levels provide additional protection of crop yield loss. Furthermore, there are specific economically significant crops that may not be protected at 15 ppm-hrs but that would be protected at lower levels. Based on scientific judgment of CASAC, a level of 10 ppm-hrs is required to reduce foliar injury. A level of 7 ppm-hours is protective of

> relative biomass loss for trees. Furthermore, 7 ppm-hours offers additional protection against crop yield loss and foliar injury. Therefore, 7 ppm hours is protective of ecosystem services. Thus, lower levels from the recommended range offer a greater degree of protection of more endpoints than do higher levels from the range.
>
> If, as a policy matter, the Administrator prefers to base the secondary standard on a three year averaging period for the purpose of program stability, then the level of the standard should be revised downward such that the annual level in any given year of the three year period would not exceed the scientifically recommended range of annual levels of 7 ppm-hrs to 15 ppm-hrs. For example, if in a three year period the highest annual W126 value is 15 ppm-hrs, and the lowest W126 value associated with a three year average is 13 ppm-hrs, then the appropriate level for the three year average would be 13 ppm-hrs to protect against a peak annual level of 15 ppm-hrs. The final Policy Assessment should quantify the ratio of the annual to three year average W126 values to determine what downward adjustment from the annual levels recommended here is needed if a three year form is selected.

Note that in his 2010 comments on CASAC's recommendations for the reconsidered O_3 primary and secondary standards (as ordered by the Obama Administration in 2009), Samet (2010) stated for the secondary standard

> CASAC also supports EPA's secondary ozone standard as proposed as a new cumulative, seasonal standard expressed as an annual index of the sum of weighted hourly concentrations (i.e., the W126 form), cumulated over 12 hours per day (8am to 8pm) during the consecutive 3-month period within the ozone season with the maximum index value, set as a level within the range of 7 to 15 ppm-hours. This W126 metric can be supported as an appropriate option for relating ozone exposure to vegetation responses, such as visible foliar injury and reductions in plant growth. We found the Agency's reasoning, as stated in the *Federal Register* notice of January 19, 2010, to be supported by the extensive scientific evidence considered in the last review cycle. In choosing the W126 form for the secondary standard, the Agency acknowledges the distinction between the effects of acute exposures to ozone on human health and the effects of chronic ozone exposures on welfare, namely that vegetation effects are more dependent on the cumulative exposure to, and uptake of, ozone over the course of the entire growing season (defined to be a minimum of at least three months). In this proposal, the Agency is responding to the clear need for a secondary standard that is different from the primary standard in averaging time, level and form.

In the subsections that follow, the use of the 1-year W126 versus the 3-year average of the W126 metric will be evaluated, as well as the use of the 3-year average of the 4th highest daily maximum concentration metric as a substitute for the 3-year W126 index.

1.9.2 1-Year W126 versus the 3-Year Average of the W126 Exposure Metric

In response to the Court's decision, the Administrator notes in his draft O_3 NAAQS rulemaking (Federal Register, 2020) that the proposed draft addresses the August 2019 decision by the D.C. Circuit on the secondary standard established in 2015 and issues raised. The EPA has expanded certain analyses in this review compared with those conducted in the last review, including discussion on issues raised in the remand, and provided additional explanation of rationales for proposed conclusions on these points in this review.

As noted by the Administrator (Federal Register, 2020, pages 49901 - 49902):

> The PA recognizes that the evidence that allows for specific evaluation of the predictability of growth impacts from single-year versus multiple-year average exposure estimates is quite limited. Such evidence would include multi-year studies reporting results for each year of the study, which are the most informative to the question of plant annual and cumulative responses to individual years (high and low) over multiple-year periods. The evidence is quite limited with regard to studies of O_3 effects that report seasonal observations across multi-year periods and that also include detailed hourly O_3 concentration records (to allow for derivation of exposure index values). Such a limitation contributes uncertainty and accordingly a lack of precision to our understanding of the quantitative impacts of seasonal O_3 exposure, including its year-to-year variability on tree growth and annual biomass accumulation (PA, section 4.3.4). The PA finds this uncertainty to limit our understanding of the extent to which tree biomass would be expected to appreciably differ at the end of multi-year exposures for which the overall average exposure is the same, yet for which the individual year exposures varied in different ways (e.g., as analyzed in Appendix 4D of the PA). Thus, the PA notes that the extent of any differences in tree biomass for two multi-year scenarios with the same 3-year average W126 index but differing single-year indices is not clear, including for exposures associated with O_3 concentrations that would meet the current standard (PA, section 4.3.4).
>
> One such study, which tracked exposures across six years, is available for aspen (King et al., 2005; 2013 ISA, section 9.6.3.2; ISA, Appendix 8, section 8.13.2). This study was used in a presentation of the 2013 ISA that compared the observed growth response to that predicted from the E-R function for aspen. Specifically, the observed aboveground biomass (and RBL) after each of the six growing seasons was compared to estimates derived from the aspen E-R function based on the cumulative multiple-year average seasonal W126 index values for each year (2013 ISA, section 9.6.3.2). The conclusions reached were that the agreement between the set of predictions and the Aspen FACE observations were "very close" and that "the function based on one year of growth was shown to be applicable to subsequent years" (2013 ISA, p. 9-135). The PA observes that such results indicate that when considering O_3 impacts on growing trees across multiple years, a multi-year average index yields predictions close to observed measurements across the multi-year time period (2013 ISA, section 9.6.3.2 and

Fig. 9-20; PA, Appendix 4A, section 4.A.3). The PA also includes example analyses that use biomass measurements from the multi-year study (King et al., 2005) to estimate aboveground aspen biomass over a multi-year period using the established E-R function for aspen with a constant single-year W126 index, e.g., of 17 ppm-hrs, or with varying annual W126 index values (10, 17 and 24 ppm-hrs) for which the 3-year average is 17 ppm-hrs, and that yield somewhat similar total biomass estimates after multiple years (PA, Appendix 4A, section 4A.3).

Thus, the PA finds that, while the E-R functions are based on strong evidence of seasonal and cumulative seasonal O_3 exposure reducing tree growth, and while they provide for quantitative characterization of the extent of such effects across O_3 exposure levels of appreciable magnitude, there is uncertainty associated with the resulting RBL predictions. Further, the current evidence does not indicate single-year seasonal exposure in combination with the established E-R functions to be a better predictor of RBL than a seasonal exposure based on a multi-year average, or *vice versa* (Appendix 4A, section 4A.3.1). Rather, associated uncertainty contributes or implies an imprecision or inexactitude in the resulting predictions, particularly for the lower W126 index estimates of interest in this review. In light of this, the current evidence does not support concluding there to be an appreciable difference in the effect of three years of exposure held at 17 ppm-hrs compared to a 3-year exposure that averaged 17 ppm-hrs yet varied by 5 to 10 ppm (e.g., 7 ppm-hrs) from 17 ppm-hrs in any of the three years for tree RBL over such multiple-year periods. The PA considered all of the factors identified here, the currently available evidence and recognized limitations, variability and uncertainties, to contribute uncertainty and resulting imprecision or inexactitude to RBL estimates of single-year seasonal W126 index values. The PA found these considerations to indicate there to be no lesser support for use of an average seasonal W126 index derived from multiple years (with their representation of variability in environmental factors), such as for a 3-year period, for estimating median RBL using the established E-R functions than for use of a single-year index.

In the Draft Review of the Ozone National Ambient Air Quality Standards document (Federal Register, 2020, page 49902), the Administrator described the results using the King et al. (2005) data as follows:

This example, while simplistic in nature, and with inherent uncertainties, including with regard to broad interpretation given the reliance on data available for the single study, quantitatively illustrates potential differences in growth impacts of W126 index, as a 3-year average, for which individual year values vary while still meeting the value specified for the average, from such impacts from exposure controlled to the same W126 index value annually. The PA suggests that this example indicates based on the magnitude of variation documented for annual W126 index values occurring under the current standard, a quite small magnitude of differences in tree biomass between single-year and

> multi-year average approaches to controlling cumulative exposure (PA, Appendix 4A, section 4A.3).

Thus, as indicated above, the PA (EPA, 2020b) appears to draw the conclusion, based on very limited data, that the current evidence does not indicate single-year seasonal exposure in combination with the established E-R functions to be a better predictor of RBL than a seasonal exposure based on a multi-year average, or *vice versa*. However, the conclusions reached in the PA using the King et al. (2005) data have limited application for clarifying differences between 1-year and 3-year W126 exposures as noted in the PA (EPA, 2020b, page 4A-23):

> Additionally, this analysis is based on aspen, and the specific pattern of differences between the two scenarios might be expected to vary for species with different biomass growth rates (and E-R functions). However, datasets of tree growth across multiple-year periods such as that available for aspen in the study by King et al., 2005) are not prevalent.

The simple question is" "How applicable are the results using the aspen data for drawing conclusions concerning the remaining 10 tree species?" The response is that it is not possible to answer this question without additional published data on other tree species. In addition, even if additional data were available, it is important to compare the final results of these types of analyses with the current state of knowledge about the importance of the higher hourly average O_3 concentrations in relation to the mid- and low-level values for assessing vegetation effects.

As indicated, the Court noted that the EPA did not explain in its 2015 decision on the O_3 NAAQS why it was reasonable to focus on a 3-year average of the W126 index instead of the 1-year W126 value recommended by CASAC. As indicated above, the PA (EPA, 2020b) appears to draw the conclusion, based on very limited data, that the current evidence does not indicate single-year seasonal exposure in combination with the established E-R functions to be a better predictor of RBL than a seasonal exposure based on a multi-year average.

As noted by the Administrator above

> In light of this, the current evidence does not support concluding there to be an appreciable difference in the effect of three years of exposure held at 17 ppm-hrs compared to a 3-year exposure that averaged 17 ppm-hrs yet varied by 5 to 10 ppm (e.g., 7 ppm-hrs) from 17 ppm-hrs in any of the three years for tree RBL over such multiple-year periods.

The EPA cautions the broad interpretation of its conclusions based on data available from a single study. If EPA were to accept the broad interpretation that no appreciable difference is observed in the effect of three years of a constant annual exposure held at 17 ppm-hrs compared to a 3-year exposure that averaged 17 ppm-hrs yet varied by 5 to 10 ppm, then it has to conclude that the distribution of hourly average O_3 concentrations for any given year (including the frequency of the higher part of the distribution) does not play an important role in predicting tree seedling growth. Thus, the Agency would apparently conclude, as long as the 3-year average of the W126 values were the same, no difference in the predicted vegetation effects would occur

even if one exposure regime over 3 years experienced many high and low hourly average concentrations, while another regime contained many mid-level hourly average concentrations and infrequent high and low hourly values. The acceptance by the Agency of the use a 3-year average of the W126 metric appears to contradict EPA's own state-of-the-science conclusion (EPA, 2020a) that the higher hourly average O_3 concentrations, which the W126 index weights more than the mid- and low values, should be provided greater weight than the mid- and low-level values.

Vegetation research results published in the literature, as well as summarized in the EPA ISA (2020a) and PA (2020b), clearly point to the importance of the higher hourly average concentrations. Musselman et al. (1983) and Hogsett et al. (1985) performed research experiments that showed that the higher hourly average concentrations were more important than the mid- and low-level values. Following their published results, a series of controlled experiments was undertaken worldwide for assessing the importance of the higher O_3 concentrations in eliciting adverse vegetation effects. These controlled fumigation experimental results provided additional evidence for emphasizing the importance of the higher concentrations in comparison to the mid- and low-level values (e.g., EPA, 1986, 1992, 1996a, 1996b, 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).

Based on the vegetation results published over the years, the EPA in its 2020 ISA (EPA, 2020a, page 8-180) concluded (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, over using indices based on mean and other exposure indices.

In 1985, based on the experimental findings that the higher hourly average O_3 concentrations were more important than the mid-and low-level values, I created the W126 exposure index. The W126 exposure index is a non-threshold metric that is described as the sigmoidally weighted sum of all hourly O_3 values observed during a specified daily and seasonal time window, where each hourly O_3 value is given a weight that increases from zero to one with increasing value. Lefohn and Runeckles (1987) proposed the use of a sigmoidally weighted index for assessing vegetation based on evidence indicating the greater relative importance of higher concentrations in affecting vegetation in comparison to the mid and lower values (Musselman et al., 1983). Lefohn et al. (1988) mathematically described and applied the W126 exposure index to develop exposure-response relationships. The W126 exposure index has the form: $W126 = \Sigma\, w_i \times C_i$ with weight $w_i = 1/[1 + M \times exp\,(-A \times C_i/1000)]$, where $M = 4403$, $A = 126$, and where C_i is the hourly average O_3 concentration in units of ppb. The M and A constants were derived based on the desire to weight the hourly average levels (1) at a value of one at ≥ 100 ppb and (2) at extremely low values below 40 ppb. The low weighting at levels below 40 ppb assumed at the time that hourly average background O_3 concentrations were mostly associated with levels below this value (EPA, 2006). As is recognized today, hourly average concentrations associated with background O_3 can, at limited times and locations, be

significantly higher as a result of stratospheric-tropospheric transport to the surface (Lefohn et al., 2011, 2012, 2014; Emery et al., 2012; Lin et al., 2012; Federal Register, 2015; EPA, 2020b). The W126 weightings for hourly average values are shown in Fig. 1-10.

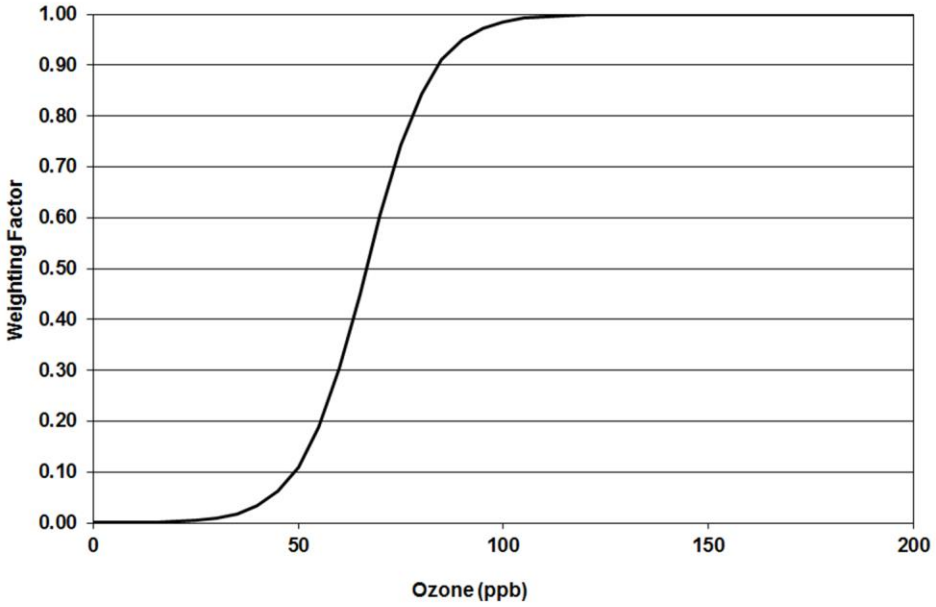


Figure 1-10. The weighting applied to hourly average ozone values for the calculation of the W126 exposure index (see Lefohn et al., 1988).

The W126 exposure index has played an important role over the past several years in assessing vegetation effects. The exposure-response relationships developed for the agricultural crop and tree seedling estimates are based on the experimental data reported in the literature. The W126 exposure-response relationships reflect the hourly O_3 concentrations used in the crop and tree growth experiments. Based on the performance of the W126 exposure metric in the exposure-response models using the experimental data, the EPA (Federal Register, 2020, page 4-34) focused its analyses in the 2008 and 2015 reviews on the W126 metric. The W126 exposure index was proposed as the O_3 secondary standard by either CASAC or EPA since 2006 three separate times (i.e., 2006, 2010, and 2014). As indicated earlier, the approach to characterize O_3 exposure concentrations for assessing potential vegetation effects, particularly growth, has received strong support from CASAC in previous reviews (Henderson, 2006; Samet, 2010; Frey, 2014). In its most current review (Cox, 2020), CASAC has again provided strong support for the W126 exposure metric.

As indicated in the ISA (EPA, 2020a), experimental evidence supports the weighting of the higher concentrations, while including the less-weighted mid and low levels. The result of this observation is that long-term average concentrations are inappropriate exposure metrics to use in vegetation exposure-response relationships. The use of a long-term average exposure metric implies that all concentrations should be treated equally, which contradicts the experimental vegetation results, as well as the empirical observations observed for vegetation in the San Bernardino National Forest. The 2013 ISA (EPA, 2013) 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 O_3 concentrations was little changed over this period. EPA (2013) suggested it was the reduction in the higher hourly average O_3 concentrations that was responsible for the improvement in tree health.

The broad interpretation of predicting tree seedling growth effects using a 3-year average of the W126 value, versus a 1-year W126, implies that the distribution of hourly average O_3 concentrations (including the frequency of the higher part of the distribution) for any given year is not necessarily important in predicting tree seedling growth. This broad interpretation appears to contradict the vegetation literature and EPA's conclusion about the importance of the higher hourly average O_3 concentrations. The use of a 3-year average of W126 exposures results in ignoring the annual exposure variability of the higher hourly average concentrations associated with vegetation effects. Important conclusions well founded in the ISA (2020a, page 8-180) appear to be contradicted. As noted earlier, EPA concluded (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, over using indices based on mean and other exposure indices. The EPA's caution concerning the broad interpretation of its conclusions about the use of the 3-year average of the cumulative W126 exposure metric for protecting vegetation is well founded. ***Based on previous scientific studies that concluded that the frequency and magnitude of the higher hourly average concentrations play an important role for estimating vegetation effects over a growth season, one needs to seriously question whether the 3-year average of the W126 metric offers the same level of protection to vegetation as a 1-year seasonal (i.e., 3-month cumulative) W126 value. The concern is that the efficacy of the W126 is compromised when a 3-year average W126 is used.***

1.9.3 The 4th Highest Daily Maximum 8-h Concentration versus the W126 Exposure Index as the Secondary O_3 NAAQS

Over the years, CASAC has emphasized that O_3 vegetation effects are more associated with the cumulative exposure over the course of an entire growing season. Therefore, the cumulative W126 exposure index provides a more biologically relevant metric for assessing vegetation risks compared to the acute exposure metric (i.e., 3-year average of the 4th highest 8-h average) associated with human health effects. In his 2010 comments on CASAC's recommendations for the reconsidered O_3 primary and secondary standards, Samet (2010) noted

> …In choosing the W126 form for the secondary standard, the Agency acknowledges the distinction between the effects of acute exposures to ozone on human health and the effects of chronic ozone exposures on welfare, namely that vegetation effects are more dependent on the cumulative exposure to, and uptake of, ozone over the course of the entire growing season (defined to be a minimum of at least three months). In this proposal, the Agency is responding to the clear

need for a secondary standard that is different from the primary standard in averaging time, level and form.

In 2010, in the reconsideration of the O_3 welfare standard, the EPA proposed to establish a distinct cumulative, seasonal "secondary" standard, referred to as the W126 index, which was designed to protect sensitive vegetation and ecosystems, including forests, parks, wildlife refuges, and wilderness areas. The proposed revisions resulted from a reconsideration of the identical primary and secondary O_3 standards set at 0.075 ppm in March 2008. In 2010, the EPA proposed to set the level of the W126 secondary standard within the range of 7-15 ppm-hours. In 2011, President Obama requested that the EPA withdraw its reconsideration of the O_3 standards, which included the secondary O_3 standard.

Following the withdrawal of the reconsideration of the O_3 standards, the EPA initiated its normal review cycle of the O_3 NAAQS. In its 2015 O_3 NAAQS decision, the Administrator chose not to select the W126 index for the secondary welfare standard. The Agency found that O_3 exposure levels associated with the existing form and averaging time were "highly correlated" to a 3-year average of the W126 index. In its August 2019 decision, the Court was concerned about the reasons the EPA chose to use the 3-year average of the 4th highest daily maximum 8-h concentration as a substitute for the W126 exposure index. The Court noted that 2014 CASAC had recommended that EPA use the 1-year W126 index as the form and averaging time for the secondary standard. The EPA had argued that adopting the W126 index as the form and averaging time was unnecessary because the 3-year average of the 4th highest daily maximum 8-h concentration was highly correlated with the 3-year average of the W126 index, especially at lower levels, and future control programs designed to help meet a primary O_3 standard based on the current form and averaging time should provide similar improvements in terms of the 3-year average of the annual W126 metric. The Court found that it lacked any basis to assess the reasonableness of EPA's actions because EPA never explained why it was reasonable to focus on a 3-year average of the W126 index instead of the 1-year W126 metric. The Court noted that the Environmental Petitioners had argued that EPA did not justify its decision not to adopt the W126 index as the form and averaging time. Therefore, the Court concluded that it could not assess the relevance of the claim that the 3-year year average of the 4th highest daily maximum 8-h concentration and the 3-year average of the W126 indices are highly correlated until the Agency had responded to why it had focused on a 3-year averaging time for the W126 index instead of the 1-year W126 metric. As noted in Section 1.9.2, based on the published literature over the past 40 years describing the importance of the higher hourly average O_3 concentrations, it is questionable whether the 3-year average of the W126 metric offers the same level of protection to vegetation as the 1-year W126 value.

Although the EPA in the PA (EPA, 2020b) devoted a considerable amount of time showing the statistical relationship between the W126 metric and the 3-year average of the 4th highest 8-h average maximum values, CASAC (Frey, 2014) noted that

> … the correlative similarity between the current standard and a level of the W126 index of 15 ppm-hrs must not be interpreted to mean that just meeting the current standard is equivalent to just meeting a W126 level of 15 ppm-hrs…

The 2014 CASAC (Frey, 2014) understood that correlative similarity did not mean that the current form of the standard could be substituted for the biologically relevant W126 for the protection of vegetation. The exposure-response relationships developed for the agricultural crop and tree seedling estimates described in the ISA (EPA, 2020a) and PA (EPA, 2020b) are based on the experimental data reported in the literature. The W126 exposure-response relationships developed from the experimental data reflect the unique patterns of hourly average O_3 concentrations applied in the crop and tree growth experiments. Vegetation researchers who developed the exposure-response models realized that the use of an average concentration, such as the 4th highest daily maximum 8-h average concentration, reduced the ability to predict the cumulative effects associated with the patterns of the hourly average concentrations used in the experimental exposure regimes. The use of exposure indices that average hourly O_3 values (e.g., the annual 4th highest daily maximum 8-h average concentration) compromises the ability to apply those exposure-response relationships, which are based on experimental hourly exposures, to ambient concentrations recorded at O_3 monitors across the U.S. for assessing vegetation effects. The 3-year average of the 4th highest daily maximum 8-h average concentration metric is not a cumulative index and therefore, will not provide adequate information to protect against those regimes that elicit adverse vegetation effects.

The Administrator, in the Draft Review of the Ozone National Ambient Air Quality Standards document (Federal Register, 2020), focused on the Court's concern about the substitution of the W126 index with the 3-year average of the annual 4th highest daily maximum 8-h concentration. The Administrator attempts to make the case that the two indices are related. Using air quality data for the 2016-2018 period, the EPA compared the W126 exposure index with the 3-year average of the 4th highest daily maximum 8-h concentration. In the PA (EPA, 2020b, page – 4D-7), the Agency notes that Fig. 1-11 illustrates a map of the observed 3-year average of the W126 metric values based on 2016-2018 data. The 3-year W126 metric values are generally at or below 13 ppm-hrs in the eastern and northwestern U.S. As noted by the Agency, the highest 3-year average W126 metric values occur in the southwestern U.S., where there are numerous monitoring sites with W126 metric values above 17 ppm-hrs.

In the EPA Draft Review of the Ozone National Ambient Air Quality Standards document (Federal Register, 2020, page 49894), the Administrator notes:

> Cumulative exposures vary across the U.S, with the highest W126 index values for sites that met the current standard being located exclusively in Southwest and West climate regions (PA, Figure 4-6). At sites meeting the current standard in all other NOAA climate regions, W126 index values, averaged over the 3-year design value period are at or below 13 ppm-hrs (PA, Figure 4-6 and Appendix 4D, Figure 4D-2). At Southwest and West region sites that met the current standard, W126 index values, averaged across the 3-year design value period, are at or below 17 ppm-hrs in virtually all cases in the most recent 3-year period and across all of the seventeen 3-year periods in the full dataset evaluated (i.e., all but one site out of 147 for recent period and all but eight out of over 1,800 cases across full dataset). Across all U.S. sites with valid design values at or below 70 ppb in the full 2000 to 2018 dataset, the W126 index, averaged over three years, was at or below 17 ppm-hrs on 99.9% of all occasions, and at or below 13 ppm-

hrs on 97% of all occasions. All but one of the eight occasions when the 3-year W126 index was above 17 ppm-hrs (including the highest occasion at 19 ppm-hrs) occurred in the Southwest region during a period before 2011. The most recent occasion occurred in 2018 at a site in the West region when the 3-year average W126 index value was 18 ppm-hrs (PA, Appendix 4D, section 4D.3.2).

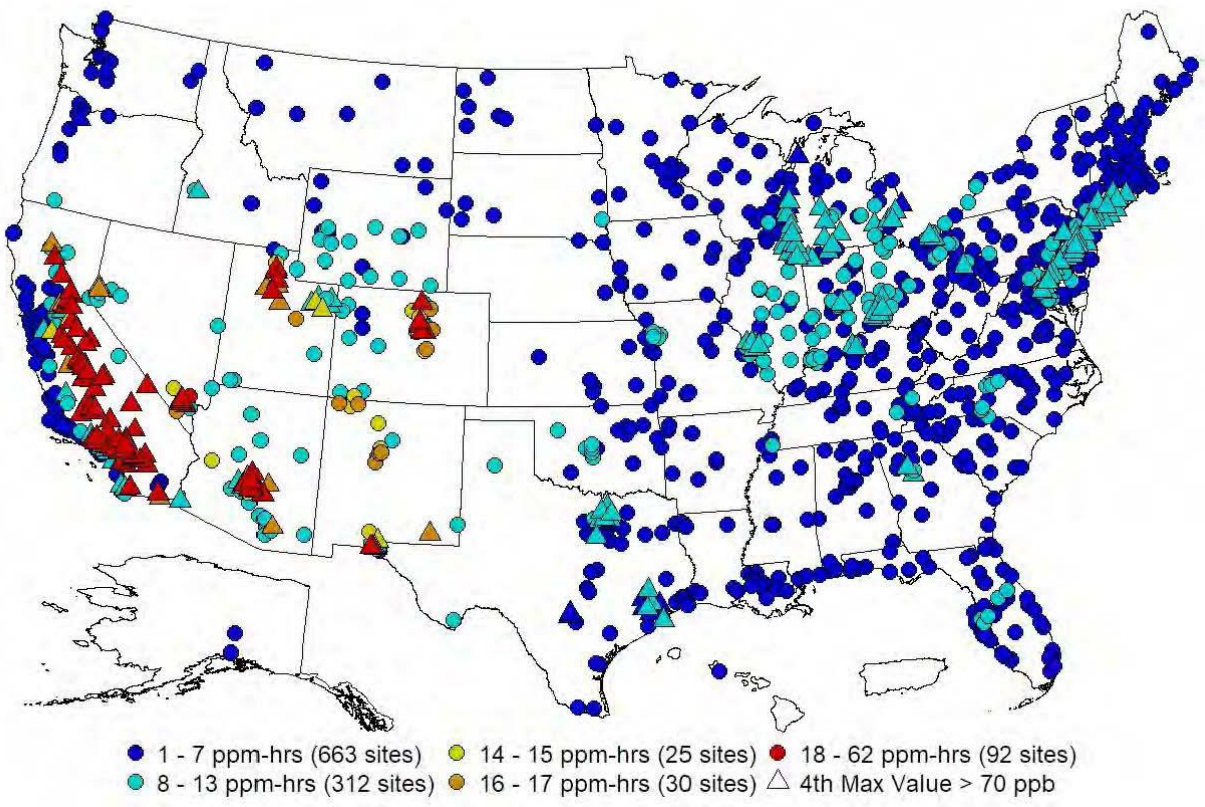


Figure 1-11. Map of W126 metric values at U.S. O_3 monitoring sites based on 2016-2018 data. Circles indicate monitoring sites with 4th max metric values less than or equal to 70 ppb, while triangles indicate monitoring sites with 4th max metric values greater than 70 ppb. Source: EPA (2020b, page 4D-6, Fig. 4D-2).

The PA (EPA, 2020b, page 4D-11) in Fig. 1-12 illustrates a scatter plot comparing the fourth-highest daily maximum 8-h concentration averaged across three years (x-axis) and 3-year W126 (y-axis) metric values based on 2016-2018 data, with points colored by NOAA climate region. This figure, according to the EPA, indicates that a strong, positive, non-linear relationship exists between the 3-year average of the 4th highest 8-h average maximum values and 3-year W126 metrics. The amount of variability in the relationship between the 3-year average of the 4th highest 8-h average maximum values and 3-year W126 metrics appears to increase as the metric values themselves increase. The relationship between the 3-year average of the 4th highest 8-h average maximum values and W126 metrics also appears to vary across

regions. In particular, as noted earlier, the Southwest and West regions (i.e., the southwestern U.S.) appear to have higher W126 metric values relative to their respective 3-year average of the 4th highest 8-h average maximum values than the remainder of the U.S.

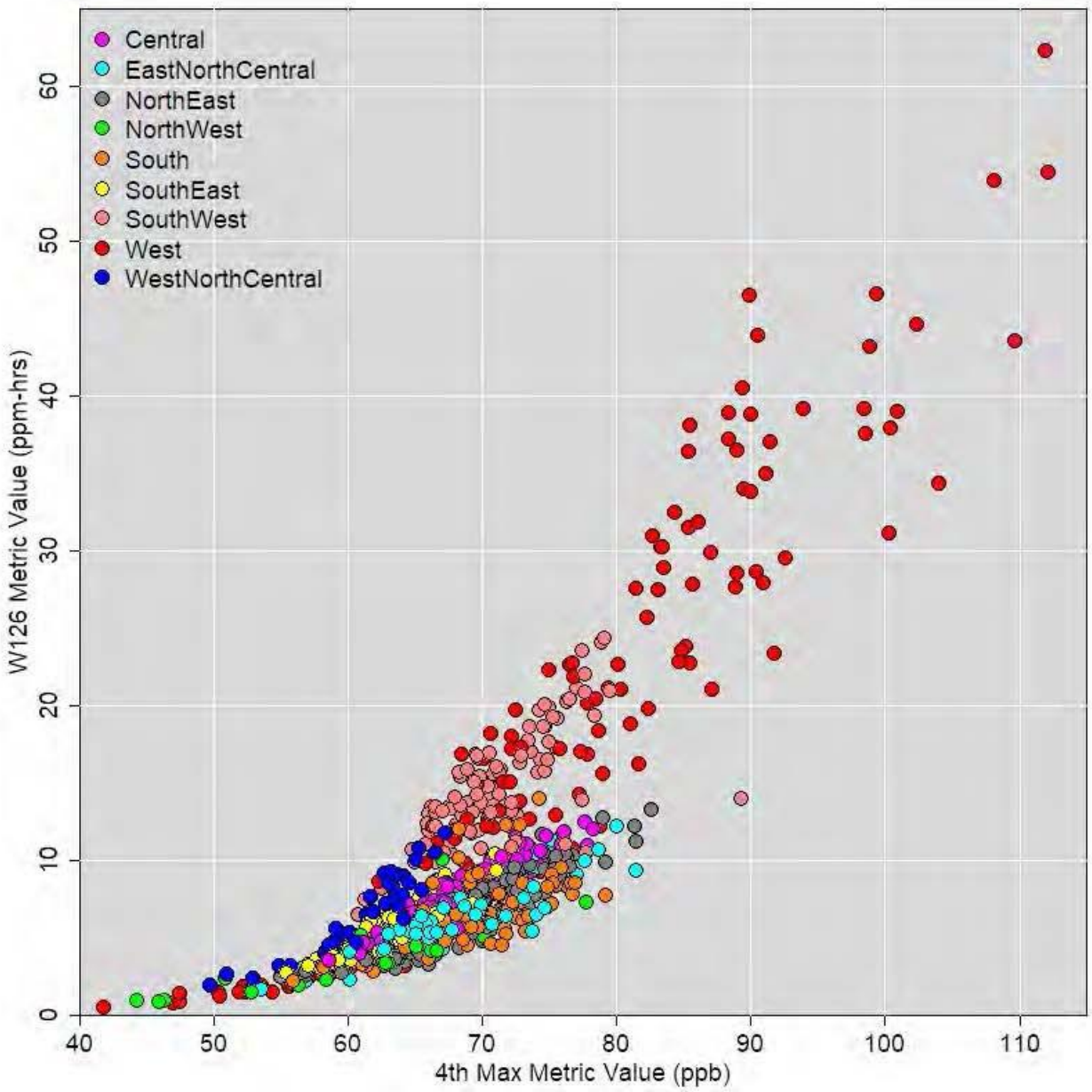


Figure 1-12. Scatter plot of W126 metric values versus 4th max metric values (design values) based on 2016-2018 monitoring data. Source: EPA (2020b, page 4D-11, Fig. 4D-3).

As noted earlier in section 1.9.2, based on limited data from the aspen results, the Administrator indicated that the current evidence does not support concluding there is an

appreciable difference in the effect of three years of exposure held at 17 ppm-hrs compared to a 3-year exposure that averaged 17 ppm-hrs yet varied by 5 to 10 ppm in any of the three years for tree RBL over such multiple-year periods. If the Agency were to draw a broad interpretation from its analyses of the King et al. (2005) data, one might conceivably conclude, if the 3-year average of the W126 values were the same at two different sites, no difference in the predicted vegetation effects would be anticipated if an exposure regime at Site 1 over 3 years experienced many high and low hourly average concentrations, while a different exposure regime at Site 2 contained many mid-level hourly average concentrations and infrequent high and low hourly values. This assumption would contradict EPA's conclusion that the higher hourly average O_3 concentrations, which the W126 index weights more than the mid- and low values, are important when assessing adverse vegetation effects (EPA, 2020a, b). The frequency and magnitude of the higher hourly average concentrations play an important role for estimating vegetation effects over a growth season.

As would be anticipated, as noted in the PA (EPA, 2020b), generally more variability in the relationship between the annual W126 index values and the 3-year average of the 4th highest daily maximum 8-h average values than the variability observed between the 3-year average of the W126 metric values and the 3-year average of the 4th highest daily maximum 8-h average values. Thus, the relationship between the 3-year W126 exposure metric and the 3-year average of the 4th highest 8-h daily maximum average concentration (Fig. 1-12) results in a "less noisier" presentation than the figure that illustrates the relationship between the annual W126 index and the 3-year average of the 4th highest daily maximum 8-h average concentration (Fig. 1-13). However, biologically, the more "noisier" figure (Fig. 1-13) provides additional insight concerning the relationship between the distribution of hourly average concentrations as indicated by the magnitude of the varying annual W126 values and the 3-year average of the 4th highest 8-h daily average maximum concentration.

The variability of the annual W126 across years is important when evaluating the ability of the 3-year average of the 4th highest 8-h daily maximum average concentration to serve as a substitute for the W126 index. Fig. 1-13 in the PA (EPA, 2020b, page 4D-13) illustrates a scatter plot comparing the 3-year average of the 4th highest 8-h average maximum values (x-axis) with the annual W126 index values (y-axis) based on 2016-2018 data, with points colored by NOAA climate regions. At a 70 ppb 3-year average for the 3-year average of the 4th highest 8-h daily maximum average concentration (Fig. 1-13), the annual W126 values range from a few ppm-hrs to 23 ppm-hrs. The large range in annual W126 values at 70 ppb implies that different exposure regimes, some with frequent occurrences of elevated hourly average levels and some with no elevated levels, would result in varying vegetation effects. Selecting the 3-year average of the 4th highest 8-h daily maximum average concentration metric does not provide an adequate index whose use can predict vegetation effects. The use of the 3-year average of the 4th highest 8-h daily maximum average concentration metric only provides EPA with a mechanism to reduce the high hourly average concentrations. The use of the 3-year average of the 4th highest 8-h daily maximum average concentration metric is not an effective index to apply to protect vegetation. EPA has recognized that O_3 effects in plants are cumulative and that adverse vegetative effects from single, high O_3 years are not offset by subsequent low O_3 years. The large variability in the annual W126 exposure index shown in Fig. 1-13 in relation to a 70 ppb 3-year average of the 4th

highest 8-h daily maximum average concentration illustrates the weakness in using the current form of the standard to adequately protect vegetation.

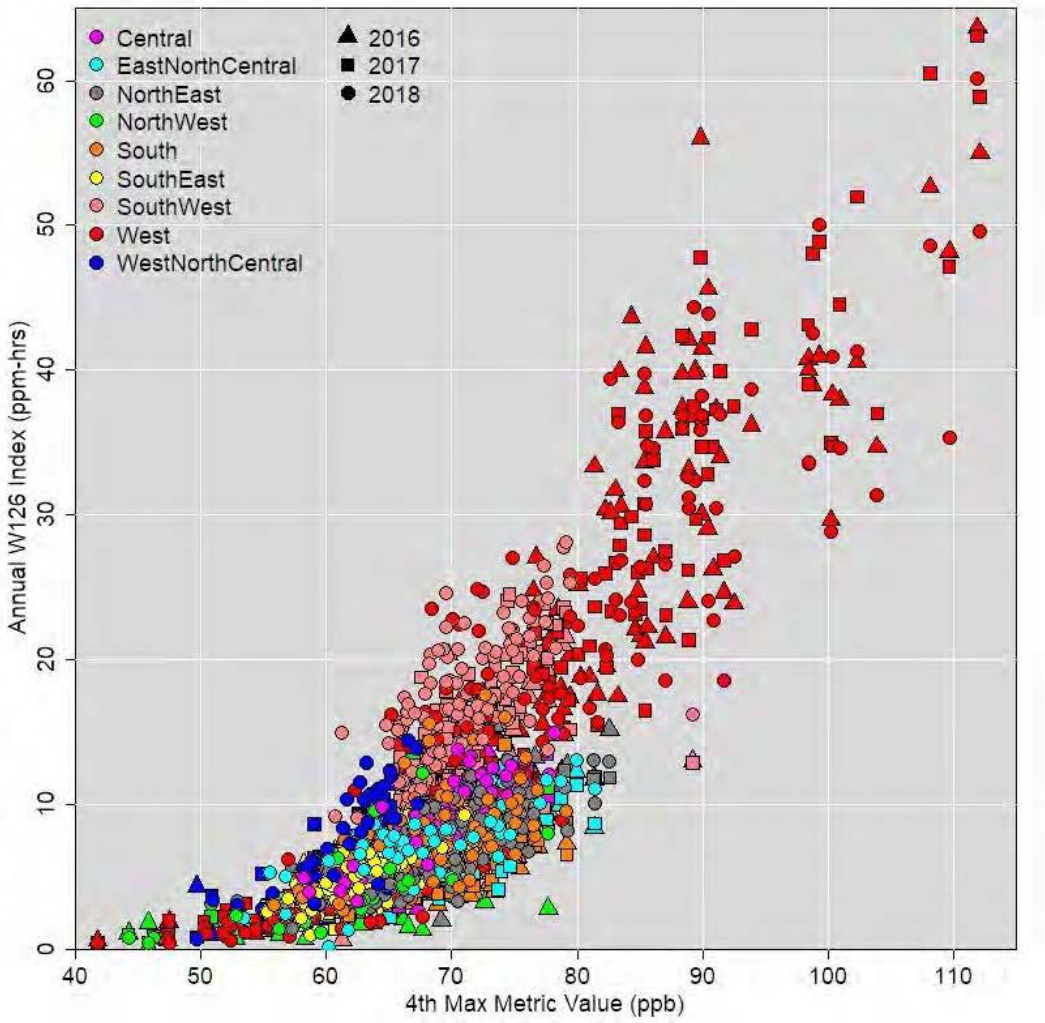


Figure 1-13. Scatter plot of annual W126 index values versus 4th max metric values (design values) based on 2016-2018 monitoring data. Source: EPA (2020b, page 4D-13, Fig. 4D-5).

In the PA (EPA, 2020b, page 2-16), Figure 1-14 (EPA, 2020b, page 2-17) illustrates a map of the site-level trends in the O_3 design values for the 3-year average of the annual 4th highest daily maximum 8-h average concentrations at 629 U.S. monitoring sites from 2000-2002 through 2016-2018. EPA notes that the design values have decreased significantly over most of the eastern U.S. during this period. Other areas of the country have also experienced decreases in design values, most notably in California and near urban areas in the intermountain west. Fig. 1-15 (EPA, 2020b, page 4D-20) in the PA is shown below. The figure illustrates the site-level trends at 638 sites in the 3-year average of the 3-month W126 metric from 2000-2002 to 2016-2018. Nearly 90% of U.S. monitoring sites experienced significant decreases in the W126 values in the eastern U.S. and California. Many locations in the western U.S. experienced little or no

change in the 3-year W126 metric over this period. The comparison of the trends in the 8-h design values with the trends in the 3-year W126 values illustrate that the two exposure metrics are behaving differently. Comparing the purple triangles, for the 8-h metric, there were 364 sites that experienced trends greater than 1 ppb/year and for the W126 metric, there were only 102 sites that experienced greater than 1 ppm-hr/year. There were 66 sites with no significant trends using the 3-year W126 metric compared to 37 sites using the 8-h metric. When comparing the trend patterns of the 3-year 8-h and W126 metrics, the two exposure metrics behave differently over time. As noted above, the trend patterns of the 3-year average of the 8-h metric were different than those of the 3-year average of the W126 index. The difference in behavior in the W126 exposure index (either annual or 3-year average) in relation to the 3-year average of the 4th highest daily maximum 8-h average concentration at 70 ppb illustrates the weakness in using the current form of the standard to adequately protect vegetation. The use of the 3-year average of the 4th highest daily maximum 8-h average concentration as a substitute for the 1-year W126 index is inadequate for protecting vegetation from those hourly average O_3 concentrations most important in eliciting adverse effects.

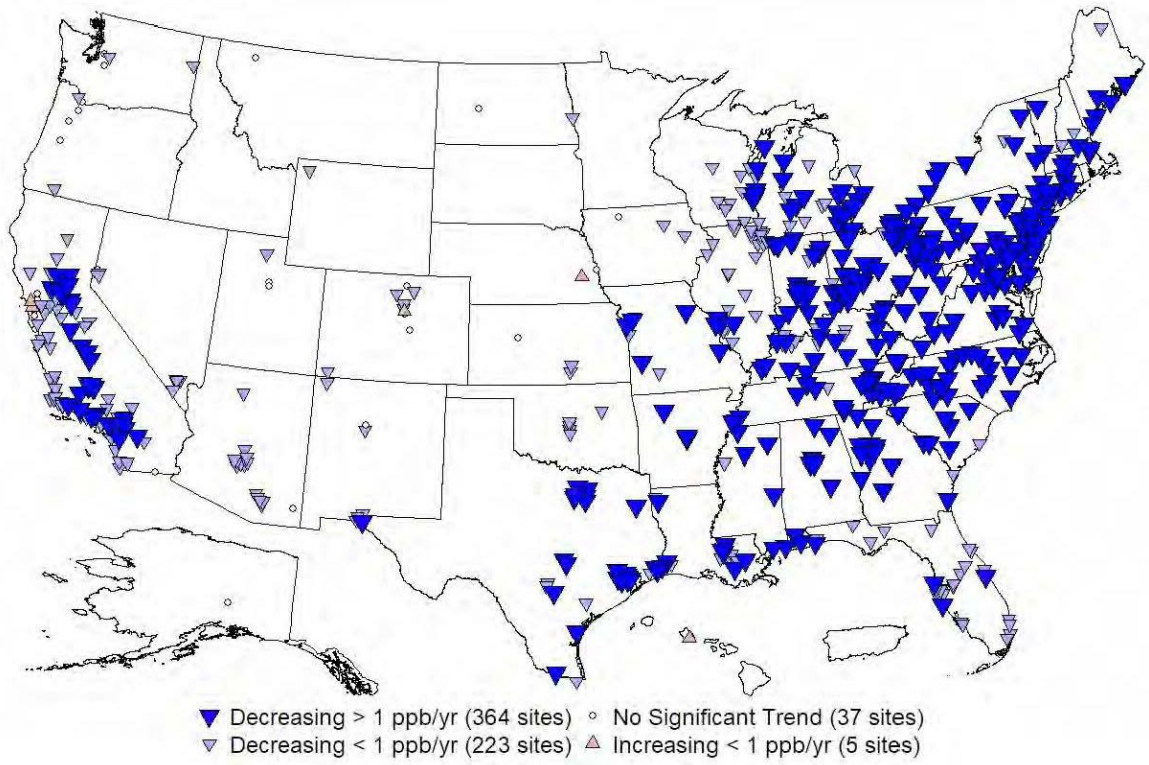


Figure 1-14. Map of trends in 3-year average of the annual 4th highest daily maximum 8-h average concentrations at U.S. O_3 monitoring sites from 2000-2002 to 2016-2018. Source: EPA (2020b, page 2-17, Fig. 2-9).

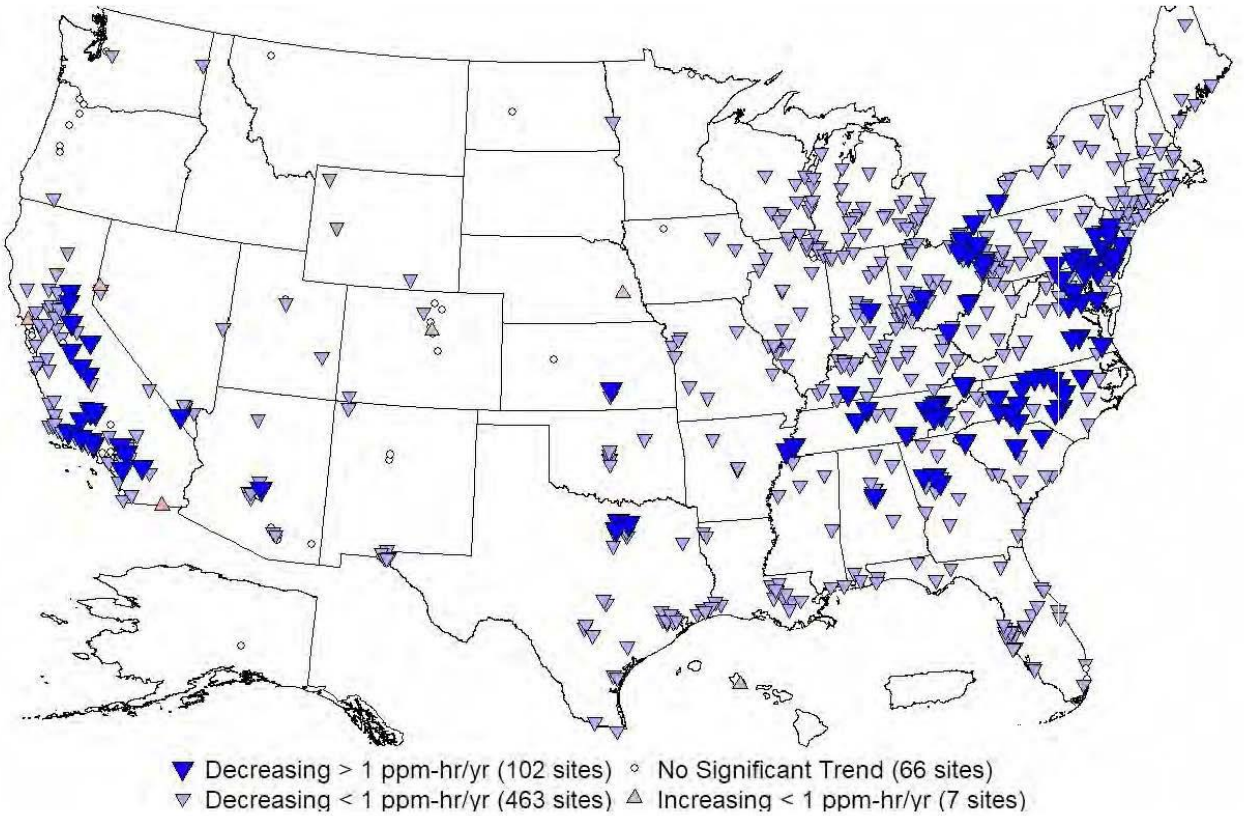


Figure 1-15. Map of trends in W126 metric values at U.S. O_3 monitoring sites from 2000-2002 to 2016-2018. Source: EPA (2020b, 4D-20, Fig. 4D-10).

As noted earlier in this subsection, it is important that exposure indices adequately capture the uniqueness of the combinations of hourly average O_3 concentrations used in experimental exposure regimes. Those concentrations that are potentially most harmful are represented according to the EPA (2020a, 2020b) by a cumulative 1-year W126 exposure index. Because the higher hourly concentrations are more important than the lower values, the use of a 3-year average of the 4th highest daily maximum 8-h average concentration will lead to a situation in which different cumulative 1-year W126 values will be observed at sites that experience the same 3-year average of the 4th highest daily maximum 8-h average concentration. In other words, the same 3-year average of the 4th highest daily maximum 8-h average concentrations (e.g., 70 ppb) can result in different distributions of hourly average concentrations and therefore, different 1-year W126 values (Fig. 1-13). This implies that while there appears to be a mathematical relationship between the current form of the human health standard and the cumulative W126 metric, the 3-year average of the 4th highest daily maximum 8-h concentration exposure index cannot adequately represent the distributions of hourly average O_3 concentrations responsible for vegetation injury and damage. In addition, because O_3 exposures vary from year to year, the process of averaging the 1-year W126 metric over a 3-year period will exacerbate the situation even more. One needs to characterize the annual W126 metric for predicting growth loss and not average across 3 years. The use of a 3-year average W126, as noted earlier, will only

introduce greater uncertainty in the predictions. As noted earlier in this discussion, CASAC's guidance in 2014 (Frey, 2014) was clear when it stated the following:

> The CASAC does not recommend the use of a three-year averaging period. We favor a single-year averaging period, which will provide more protection for annual crops and for the anticipated cumulative effects on perennial species. The scientific analyses considered in this review, and the evidence upon which they are based, are from single-year results. If a 3-year averaging period is established, then the upper limit will need to be reduced to protect against one-year ozone peaks. We consider this further in the response to charge questions for Chapter 6.

The use of the 3-year average of the 4th highest daily maximum 8-h average concentration as a substitute for the 1-year W126 index is inadequate for protecting vegetation from those hourly average O_3 concentrations most important in eliciting adverse effects. The current form of the primary standard is used as a blunt tool for a job that requires a more precise object (i.e., the W126) for solving a major task: the protection of vegetation across the U.S. Therefore, ***given the above discussion in this section and the state of knowledge, the best protection for vegetation effects associated with O_3 exposures is to adopt the 1-year W126 exposure metric as the form of the secondary standard, which is different in averaging time, level, and form of the human health 3-year average of the 4th highest daily maximum 8-h average concentration.***

2. Additional information about 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 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 ISA (EPA, 2020a, 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. In addition, Hazucha et al. (1987), based on their meta-analysis, concluded that O_3 concentration ''exerts a greater influence'' on spirometric lung function decline than the intensity of exercise (i.e., minute ventilation). Conclusions from important papers discussing the importance of the higher hourly average O_3 concentrations on human health 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., 2010). 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, 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 ISA (EPA, 2020a) notes the following:

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 reason that the FEV_1 responses were similar was because there was "recovery" occurring in the variable exposure regime versus the cumulative response based on the square-wave exposure. Following the peak FEV_1 response, usually 1 to 2 h after the peak hourly O_3 concentration, lung function improved despite continuing O_3 exposure. Results from the 80 ppb variable profiles applied in Adams (2003, 2006a) show a recovery as O_3 concentrations decline to 50 ppb. Similarly, results from Hazucha et al. (1992) and Adams (2006b) show reversal of FEV_1 response as the concentration drops from 60 to 0 ppb. The relatively rapid recovery found during the variable exposures at 20 EVR possibly reflects replenishment or enhancements of airway antioxidant levels as well as decreased stimulation of lung receptors as O_3 concentrations decrease towards 50 ppb and lower.

The observation 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 variable concentration regimes.

The hourly averaged concentrations were designed to increase in an "almost" symmetric manner. For the 80 ppb stepwise exposure, the peak hourly average concentration occurred at 4^{th} 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 was the 2 to 3-h initial "induction phase" in response. The second phase was the onset of a statistically significant FEV_1 nonlinear "response phase," and the final phase was 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 was 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 ISA (EPA, 2020a) and was presented as evidence supporting the statement on page 3-11 in 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 constant 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 appears to indicate that the 8-h average form of the O_3 standard may need to be revisited sometime in the future.

Conclusions from the Hazucha et al. (1992) and Adams (2003, 2006a) results illustrate 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, there should be varying hour-by-hour concentration regimes over the time of exposure, as well as more extensive research on the "induction" and "reversal" phases noted by previous investigators.

During the 2015 rulemaking O_3 activity, as well as the current rulemaking activity (Federal Register, 2020), 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 focused 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 Agency's rationale in the 2015 O_3 rulemaking that provided 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 current ISA (EPA, 2020a, page IS-1) again indicates that the strongest evidence comes from controlled human exposure studies demonstrating O_3-induced decreases in lung function and inflammation in healthy, exercising adults. The current PA (EPA, 2020b, page 3-93) notes that epidemiologic studies provide limited insight regarding exposure concentrations associated with health outcomes that might be expected under air quality conditions that meet the current standard.

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, had 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 and other researcher's trend 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 current ISA (EPA, 2020a, page 8-180) again concurs with EPA's conclusions in 2015 about the importance of the higher concentrations for eliciting vegetation effects.

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 with the EPA researchers, 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) published 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 was undertaken worldwide for assessing the importance of the higher O_3 concentrations in eliciting a vegetation response. These controlled fumigation experimental results provided additional evidence for emphasizing the importance of the higher concentrations in comparison to the mid- and low-level values (e.g., EPA, 1986, 1992, 1996a, 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).

Based on the experimental evidence, these experiments helped 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 a thorough review of the literature, EPA (2013, 2020a) concluded 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, over using indices based on mean and other exposure indices. These conclusions have been reiterated in the current ISA (page 8-180). The current ISA (EPA, 2020a, page 8-181) notes that no recent information available since the 2013 Ozone ISA alters these basic conclusions.

As indicated in the ISA (EPA, 2020a, page 8-181), 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 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 (Heath et al., 2009). As noted above in the ISA (EPA, 2020a), those species having high amounts of detoxification potential may, in fact, show little relationship between O_3 stomatal uptake and plant response. 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). Lefohn et al. (2018) 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 (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).

Lefohn et al. (2018) 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) 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.

The 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 EPA addressed the issue of using seasonal average concentration metrics to protect vegetation. In the early 1980s, the EPA considered 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 (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 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 (EPA, 1996; 2006; 2013; Federal Register, 2015). As mentioned 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 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, 1996a, 2006, 2013) noted in its literature reviews, both the seasonal average and SUM00 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 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, where the sigmoidal weighting is described in Lefohn and Runeckles (1987) and the W126 metric is described in 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 cutoff (i.e., 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.

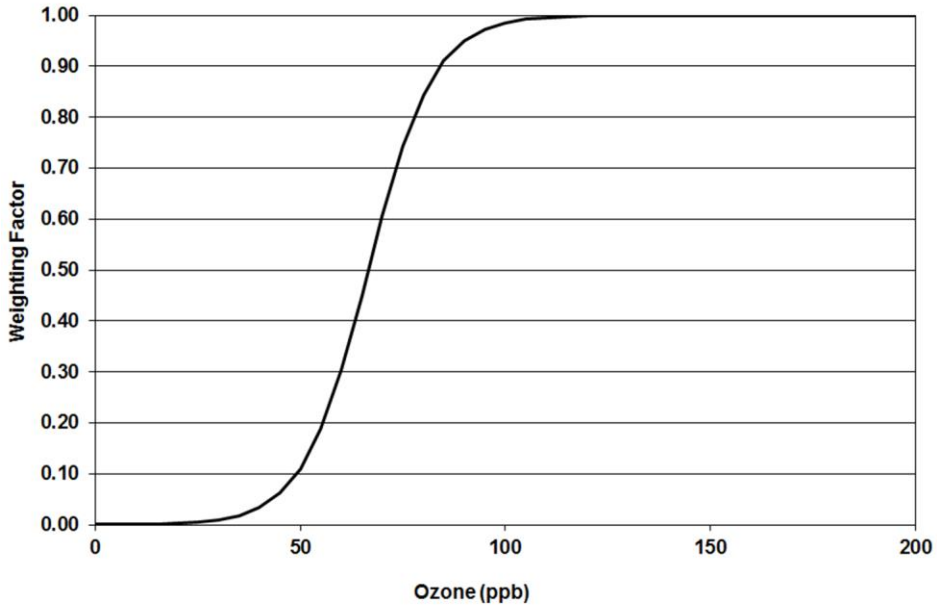


Figure 2-1. The weighting applied to hourly average O_3 values for the calculation of the W126 exposure index (see Lefohn and Runeckles, 1987 and Lefohn et al., 1988 for further details).

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 O_3 concentrations was little changed over this period. EPA (2013) suggested it was the reduction in the higher hourly average O_3 concentrations that was responsible for the improvement in tree health.

As indicated above, based on a thorough review of the vegetation literature, (1) O_3 effects in plants are cumulative; (2) higher O_3 concentrations are 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, 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 ISA (EPA, 2020a) and PA (EPA, 2020b) continue to focus on the W126 cumulative exposure index as the metric to protect vegetation. The ISA (EPA, 2020a, page 8-181) noted that McGrath et al. (2015) reported for maize and soybean yield that the SUM06, AOTx, and W126 metrics performed well and the W126 was the preferred metric because it was potentially the most sensitive index. Mills et al. (2018) noted that their results illustrated that the largest range of exposure values worldwide was for the W126 metric, where the stronger weighting for the

highest O_3 concentrations resulted in areas with the highest W126 values stand out from those with lower values. A similar, but less pronounced worldwide exposure pattern was shown for the AOT40 metric, with the M12 metric showing proportionately less spatial variation across the worldwide sites, especially in the northern hemisphere.

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 in its Air Quality Index (AQI) reporting across the U.S. (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 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-h 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, 2020) (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 used by various groups to count the number of exceedances above 70 ppb.

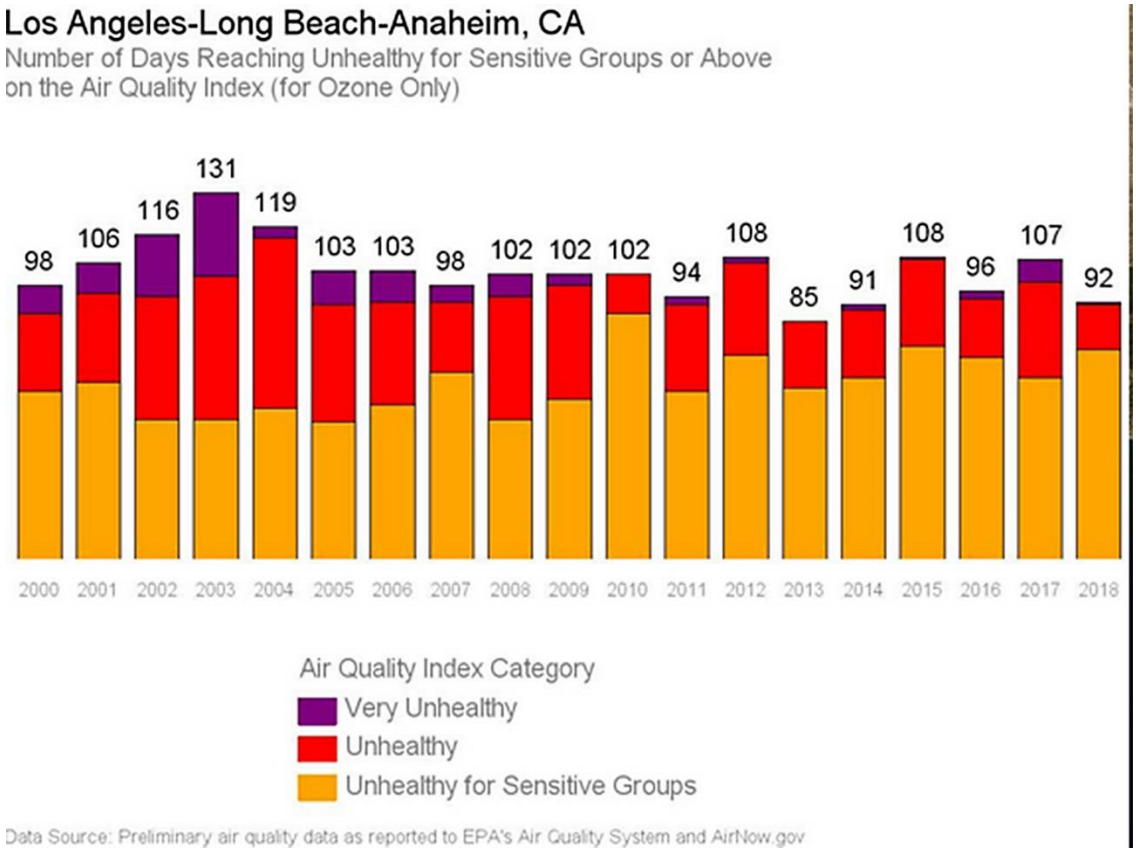


Figure 2-3. Number of days reaching unhealthy levels for sensitive groups or above on the Air Quality Index for Los Angeles-Long Beach -Anaheim, CA. Source: EPA, 2019: A Look

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 for O_3 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 quantification of the highest hourly average concentrations, which are more biological important than the lower and mid values, is lost. If the long-term average is used, then a comparison among different O_3 monitoring sites results in many of the sites having similar long-term average concentrations which 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. The relationship of the long-term average to the occurrence of the higher concentrations will be discussed further in Section 3.

Following the setting of the 2015 O_3 standard (Federal Register, 2015) and the current O_3 rulemaking activity (Federal Register, 2020), 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). **Although specific exposure metrics may appear to work in a selected effects model, what is most important is that the exposure metrics be biologically relevant and defensible**. In many cases, it may not be possible to identify a biologically relevant exposure metric based on modeling results. Prior to selecting and running a model for assessing human health or vegetation effects/risks, it is important to identify an exposure metric for the model that is justified based on biological principles.

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 U.S. sites studied in their analysis, 162 sites showed negative trends, 32 showed no trends, and 2 had insufficient data. In comparison, using the 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 PA (EPA, 2020b, page 2-20) notes that trends analyses show that metrics impacted by averaging longer time periods of hourly O_3 measurements, such as the 6-month (April-September) average of daytime (8am-7pm) O_3 concentrations, show more variation than metrics that focus on the higher concentrations. Using the same data from U.S. sites, Lefohn et al. (2017) note that for reduction emissions the trend patterns of the (1) 4th highest annual 8-h average exposure metric and (2) 6-month 12-h daylight average exposure index were extremely different and provided an excellent example of why it is so important to select appropriate biologically relevant exposure metrics 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 2-1 below provides a summary of the relationship between (1) 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 (2) median and mean values. As indicated above, overall, trends in the four mean/median metrics (i.e., median annual, mean annual, median summer, and mean summer) were not representative of the trends behavior of those metrics associated with the higher concentrations.

Table 2-1. 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 experiencing much greater exposures in China than in Japan, South Korea, Europe, and the United States, were similar in values for these other 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), 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 by reducing the higher hourly average O_3 concentrations. As emission reductions occur, the higher part of the distribution of hourly average concentrations move downwards toward the middle of the hourly average values. As a result of reducing NO_x emissions, a reduction in the NO titration of O_3 occurs with the result that lower hourly average O_3 concentrations shift upwards. This was discussed in Section 1 and will be further discussed in

Section 3. As mentioned previously in this section, as emissions are reduced, annual averages or seasonal averages tend to be related to the behavior of the upward shifts in the lower hourly average O_3 concentrations. Thus, for the purpose of assessing human health or vegetation chronic effects, one might wish to select exposure metrics based on the repeated occurrences of the higher hourly average concentrations over time. As stated previously, the EPA believed in 2015 that adequate protection for both acute and chronic exposures can be attained by focusing on the reduction of the repeated occurrences of exposures of concern (i.e., the higher hourly average concentrations). This is an especially important strategy to implement. For without the focus on the highest hourly average O_3 concentrations for reducing effects, one might believe that the increasing lower hourly average concentrations that occur during emission reductions will have a detrimental effect on human health and vegetation. However, we know that based on the scientific literature about the importance of the higher hourly average concentrations discussed in this section, the shift from the lower concentrations toward the mid-level values should not necessarily be detrimental to human health and vegetation. In its current draft proposal (Federal Register, 2020), the EPA may wish to make a clear statement repeating its 2015 conclusion that adequate protection for both acute and chronic exposures can be attained by focusing on the reduction of the repeated occurrences of exposures of concern (i.e., the higher hourly average concentrations). Without a definitive statement about the importance of the higher concentrations, the Agency may be faced with the criticism that its current O_3 control strategies are resulting in greater risk to human health and vegetation because the lower hourly average values are increasing to the mid-level concentrations in many urban areas in the U.S. Based on the scientific evidence presented in this section that supports Fundamental Principle No. 1, this criticism is unwarranted.

3 Additional information about 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

Introduction

Based on research studies and "natural experiments," the first fundamental principle discussed in Sections 1 and 2 indicate that the higher hourly average O_3 concentrations should be weighted more than middle and lower values when assessing human health and environmental effects. Emission control strategies in the U.S. focus on the reduction of the higher hourly average O_3 concentrations. In 2015, the EPA Administrator believed by reducing *the higher part of the distribution of hourly average concentrations* (not just the peak hourly values that the risk to human health and vegetation would be reduced. The EPA in its 2015 decision (Federal Register, 2015 – pages 65358 – 65359) anticipated that a revised standard with a level of 70 ppb would also reduce the occurrence of exposures to O_3 concentrations at least somewhat below 60 ppb based on its modeling results in the 2014 Health Risk and Exposure Assessment document (EPA, 2014b, Figs. 4-9 and 4-10). The modeling results illustrated that as emissions were reduced, the O3 concentrations would move downwards toward the middle hourly average O_3 values, while the lower concentrations would move upwards toward the middle values.

The observation of the lower hourly average O_3 concentrations shifting upward as emissions are reduced define the basis for Fundamental Principle No. 2. As discussed in Section 2, if one were to assume that to control for chronic health effects that annual or seasonal averages must be reduced, then chemical models (EPA, 2014b, Figs. 4-9 and 4-10), as well as empirical air quality data, indicate that annual and seasonal average metrics will not perform as some researchers anticipate. 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 (i.e., the compression effect). These patterns have been reported in the literature by various researchers (e.g., Lefohn et al., 1998; EPA, 2014b, 2020b; 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. Although the U.S. Court of Appeals for the District of Columbia Circuit (DC Circuit) in 2019 ruled that background O_3 should not directly influence the setting of the level of the NAAQS, as discussed in Section 1.5, background O_3 levels influence risk assessments associated with margin of safety considerations. 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 O_3 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 very lowest 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 is discussed in later subsections. The distribution of background O_3 hourly average concentrations will determine for each site the range of hourly average concentration values in the limit as emissions are reduced. 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. As described in Section 1.8, the severe reduction of emissions during the COVID-19 lockdown in many countries during the spring (northern hemisphere) and fall (southern hemisphere) of 2020, resulted in a "natural experiment" that provided confirmation on the behavior of hourly average O_3 concentrations at both the high end and low end of the distribution. The "natural experiment,"

as well as models and empirical data collected over the years, provide us with important insight about the behavior of changes in the distribution of O_3 concentrations.

3.1 Patterns of 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 titration of O_3 of the lower hourly average concentrations as reduction in NO_x emissions occurs (Lefohn et al., 1998; 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 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 PA (EPA, 2020b), 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 3-3 to 3-10 (reproduced from Fig. 3C-67, page 3C-103 through Fig. 3C-74, page 3C-110) 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 were 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 (Figs. 3-3 to 3-10) reproduced from the PA (EPA, 2020b) illustrate 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 PA (page 3C-101) 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).

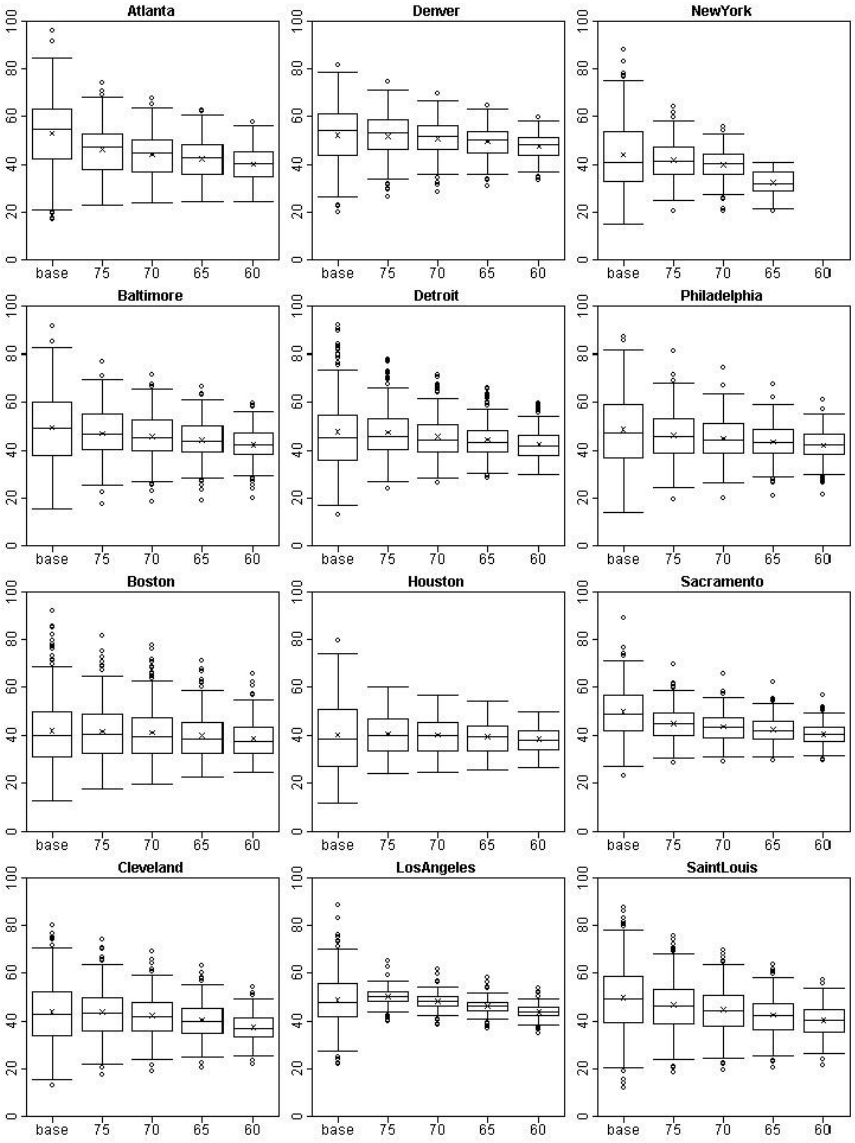


Figure 4-9. Distributions of Composite Monitor Daily Maximum 8-hr Average Values for the 12 Urban Study Areas in the Epidemiology-based Risk Assessment. Plots depict values based on ambient measurements (base), and values obtained with the HDDM adjustment methodology when just meeting the 75, 70, 65 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 EPA (2014b).

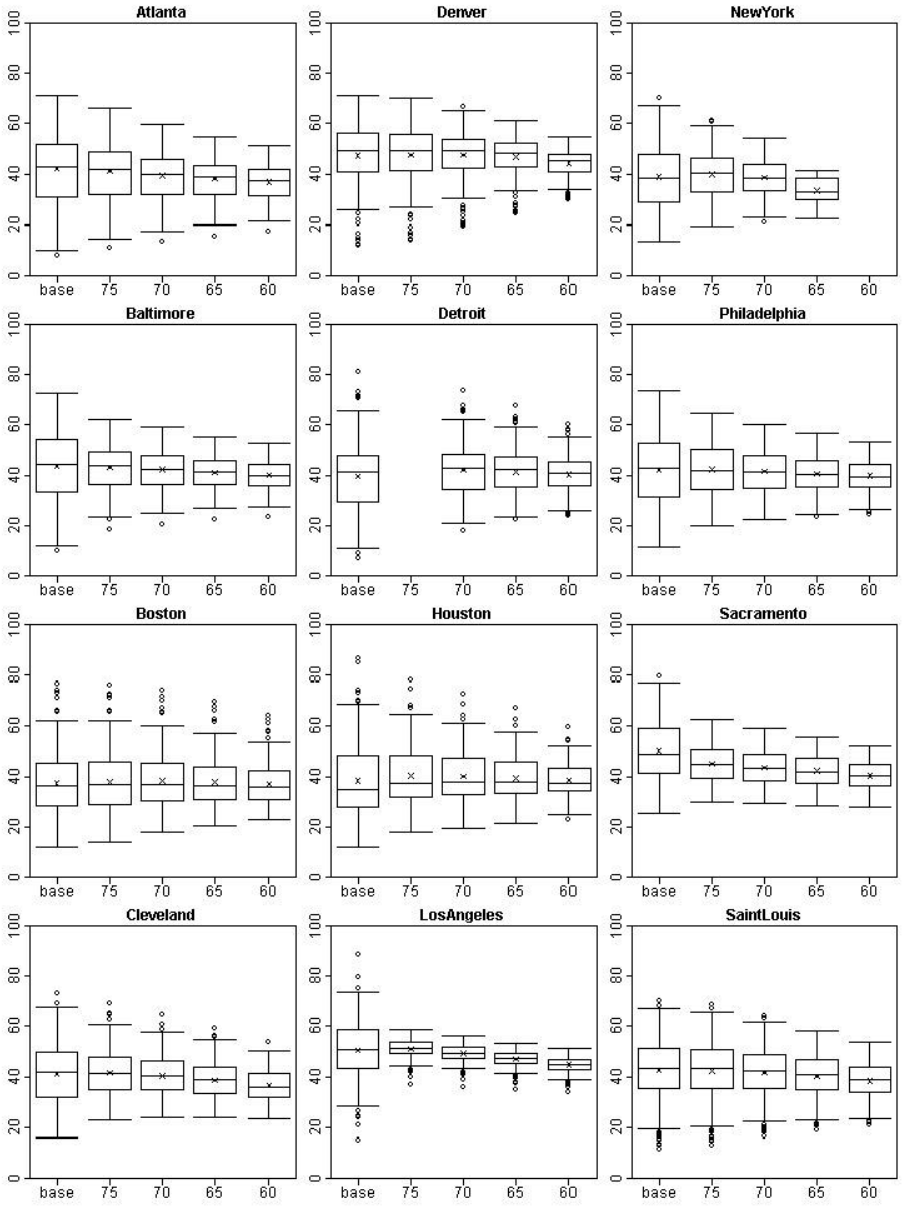


Figure 4-10. Distributions of Composite Monitor Daily Maximum 8-hr Average Values for the 12 Urban Study Areas in the Epidemiology-based Risk Assessment. Plots depict values based on ambient measurements (base), and values obtained with the HDDM adjustment methodology when just meeting the 75, 70, 65 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 EPA (2014b).

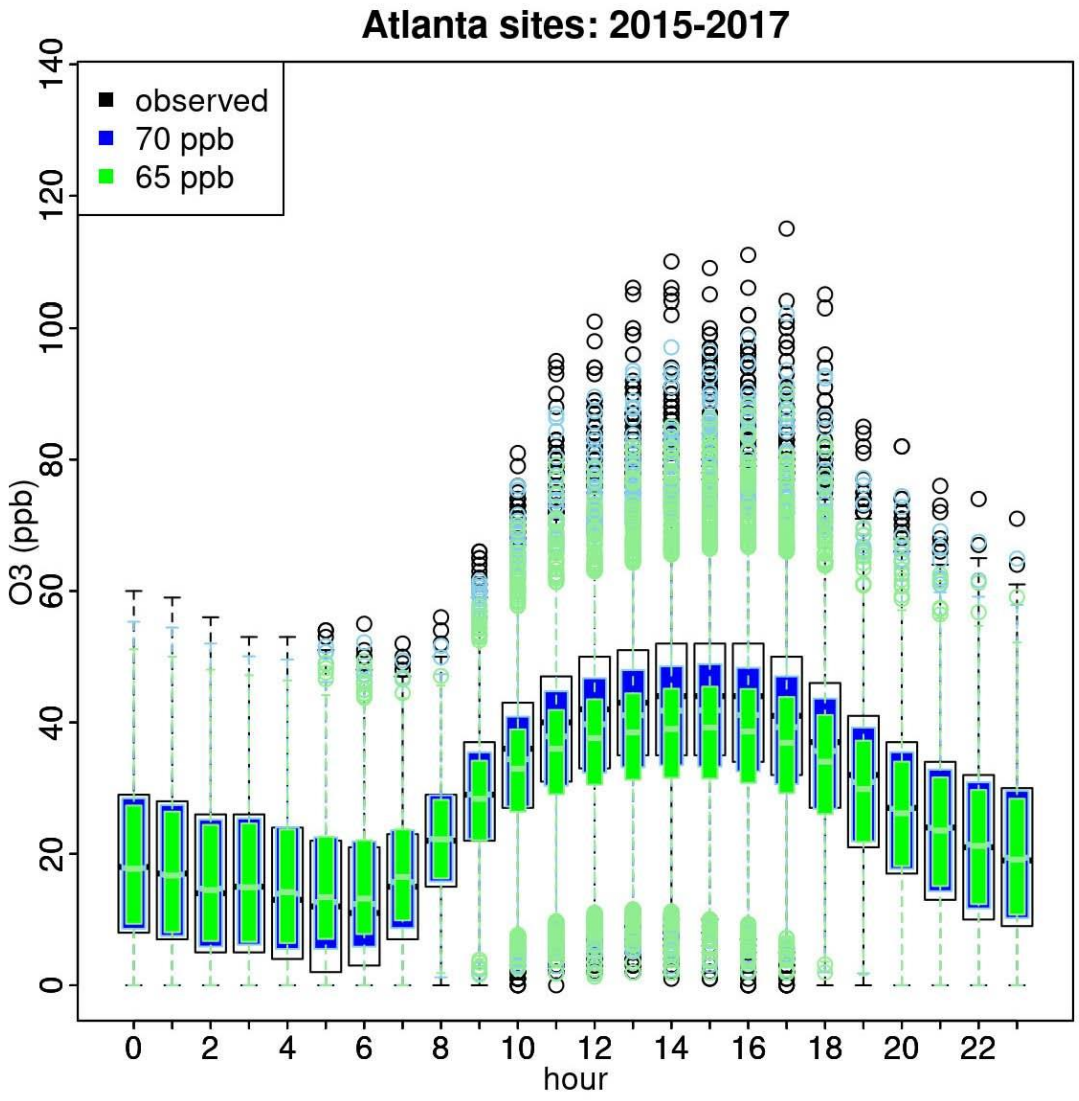


Figure 3C-67. Diurnal distribution of hourly O_3 concentrations at monitoring sites in Atlanta.

Figure 3-3. Figure 3C-67 from EPA (2020b).

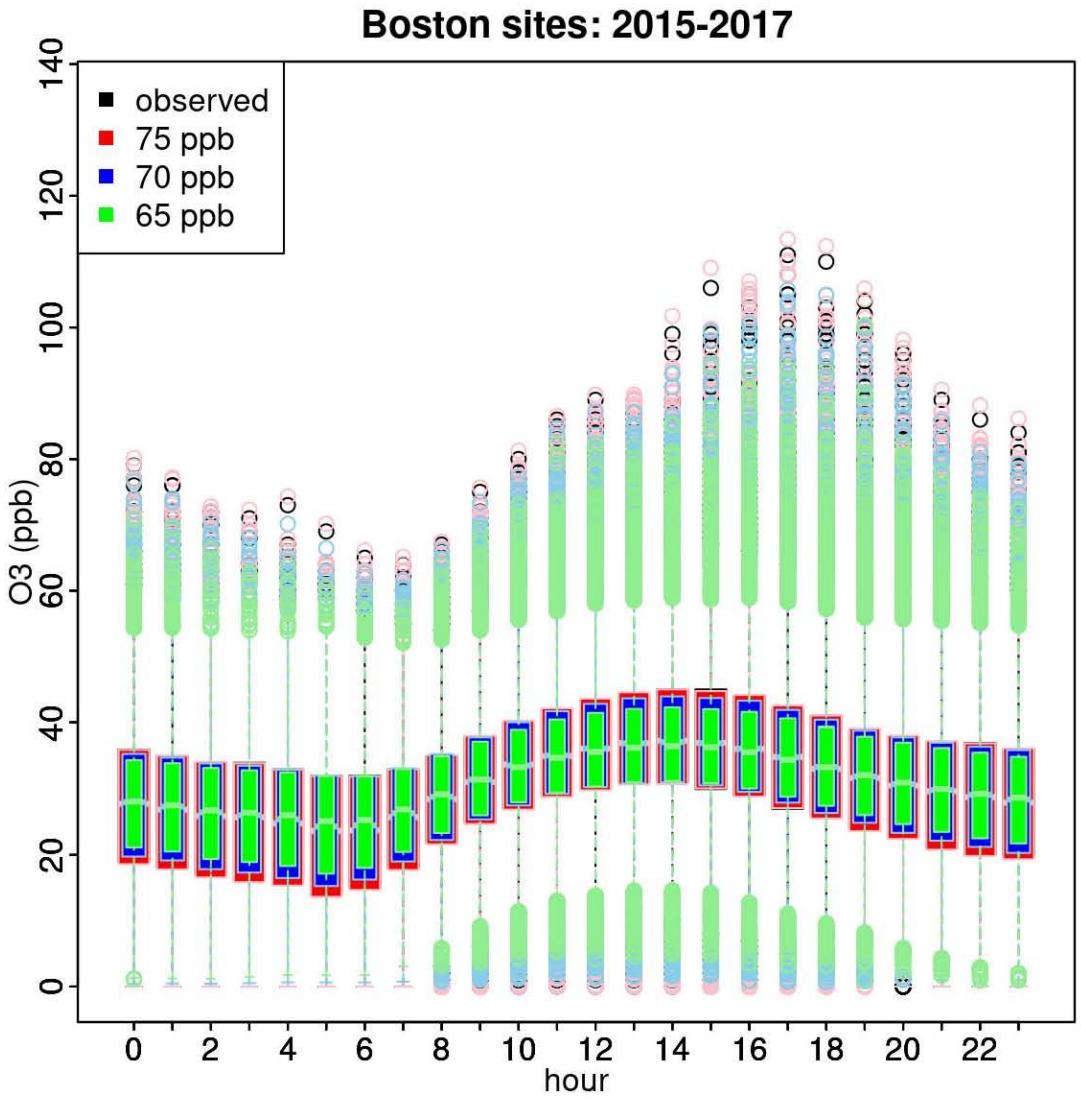


Figure 3C-68. Diurnal distribution of hourly O_3 concentrations at monitoring sites in Boston.

Figure 3-4. Figure 3C-68 from EPA (2020b).

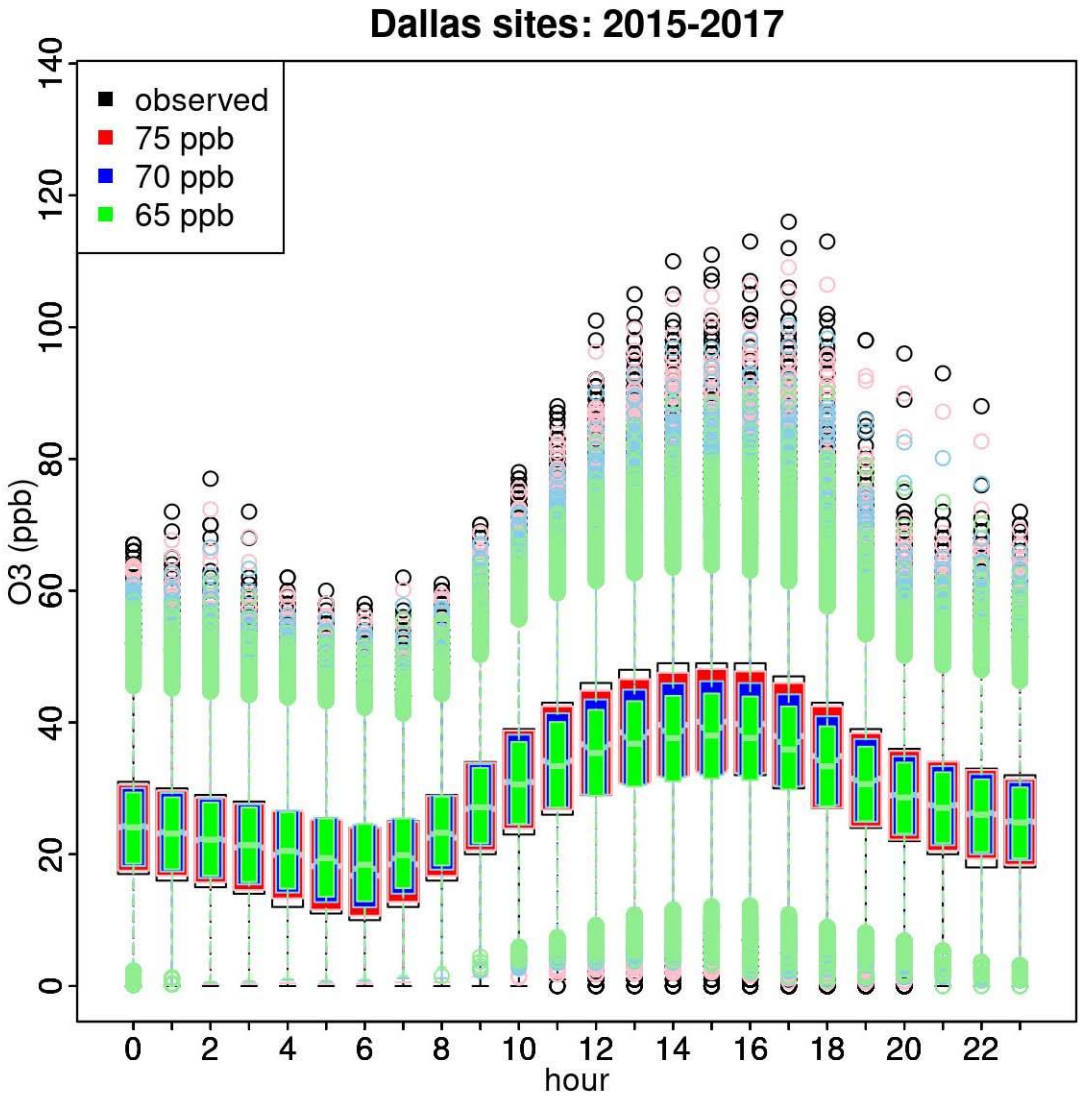


Figure 3C-69. Diurnal distribution of hourly O_3 concentrations at monitoring sites in Dallas.

Figure 3-5. Figure 3C-69 from EPA (2020b).

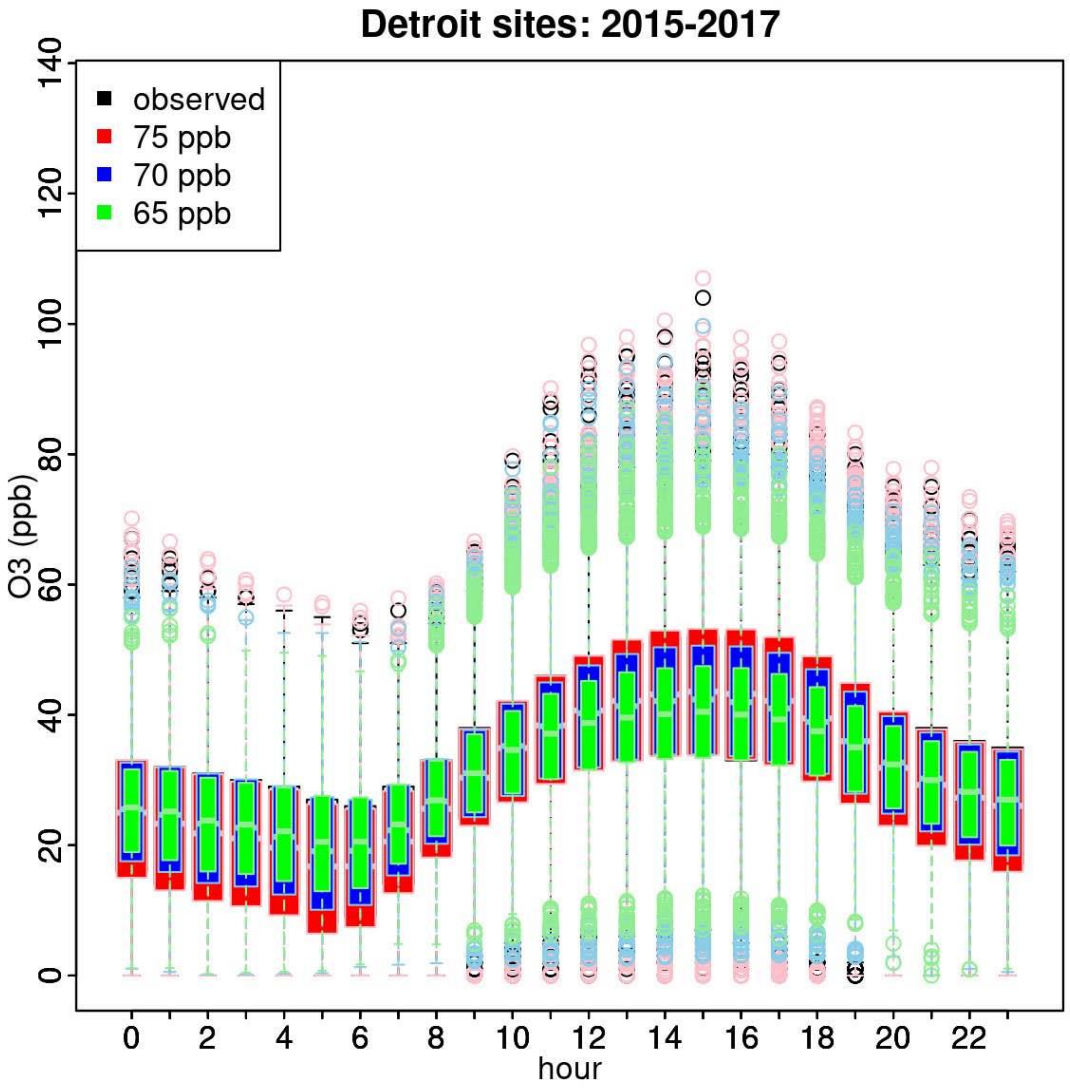


Figure 3C-70. Diurnal distribution of hourly O_3 concentrations at monitoring sites in Detroit.

Figure 3-6. Figure 3C-70 from EPA (2020b).

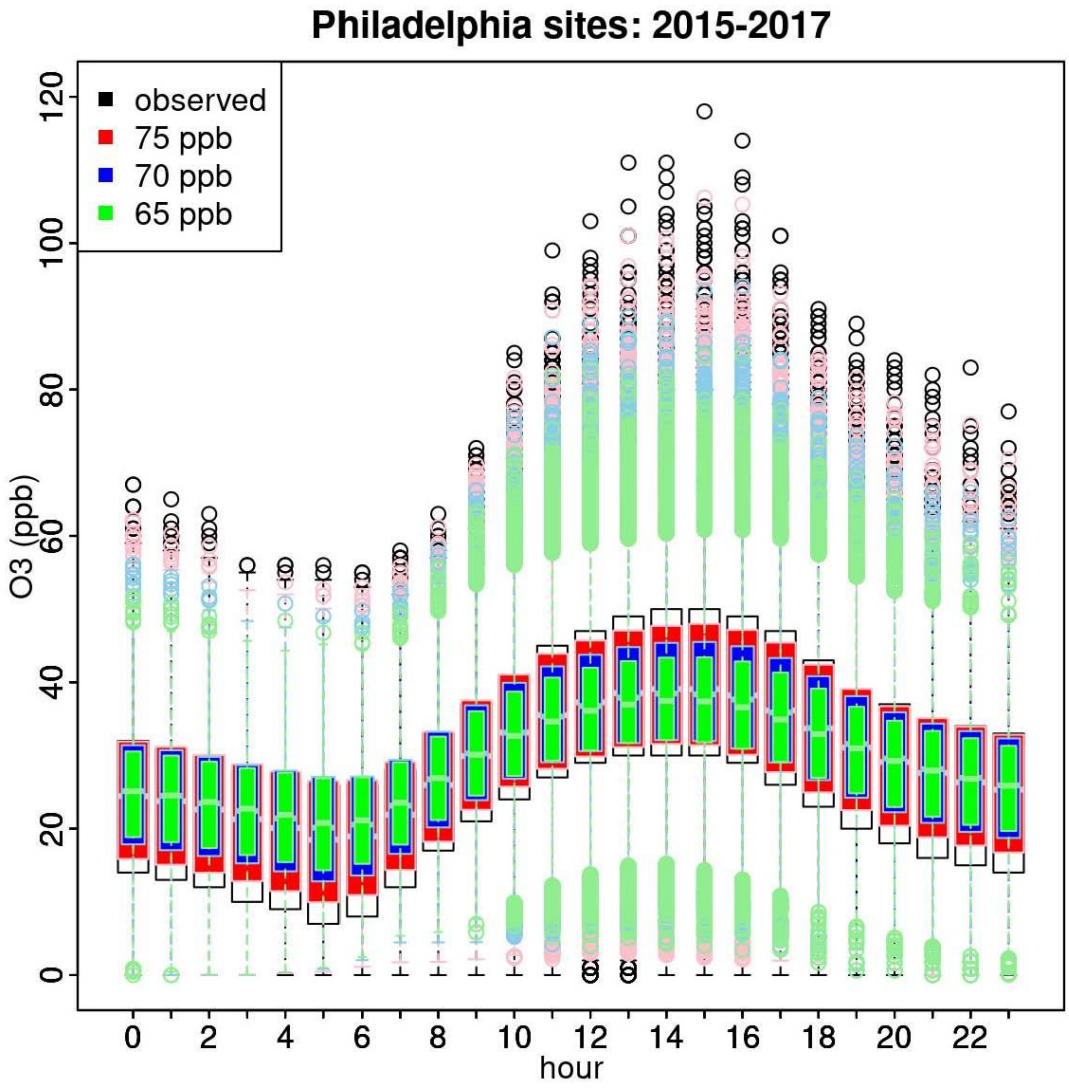


Figure 3C-71. Diurnal distribution of hourly O_3 concentrations at monitoring sites in Philadelphia.

Figure 3-7. Figure 3C-71 from EPA (2020b).

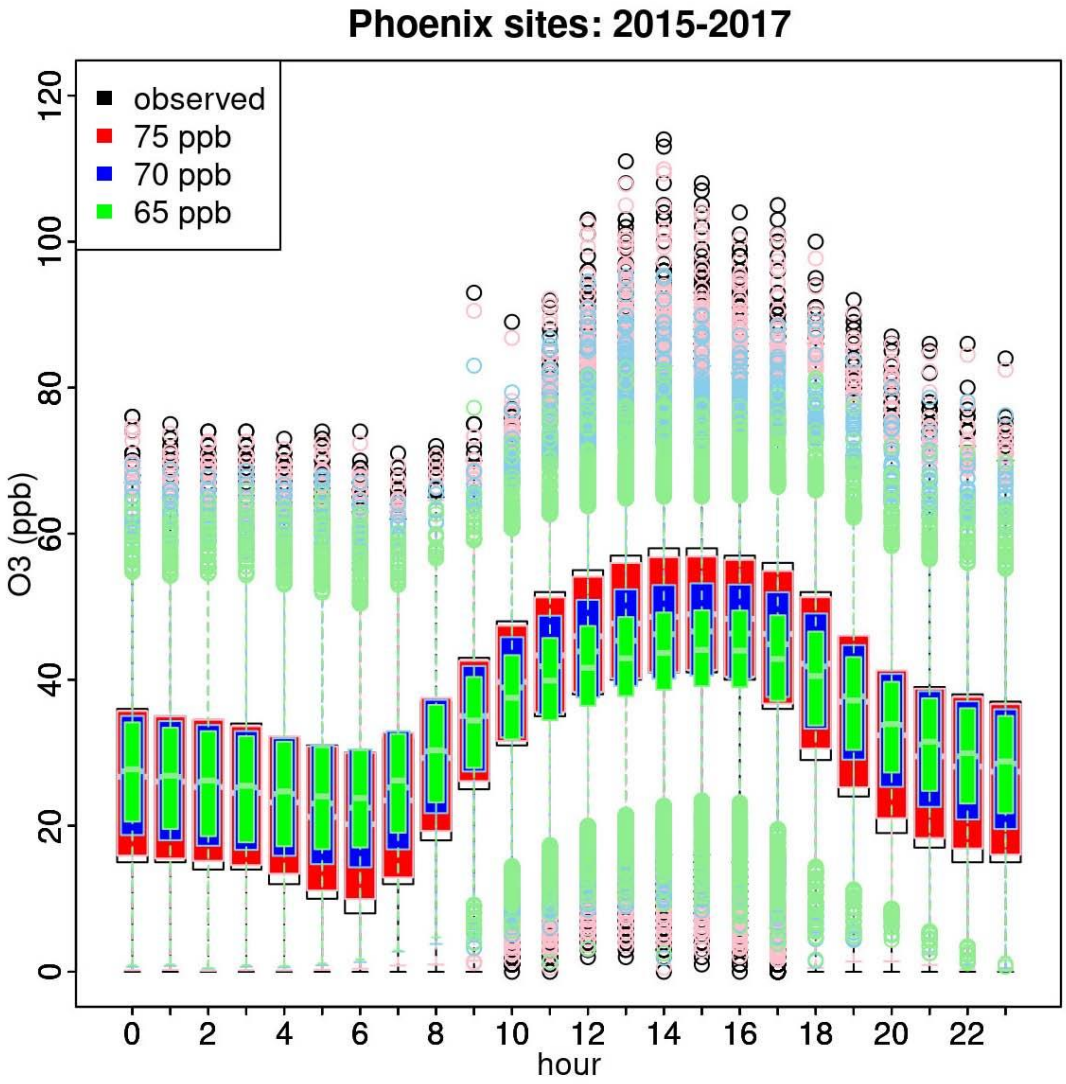


Figure 3C-72. Diurnal distribution of hourly O_3 concentrations at monitoring sites in Phoenix.

Figure 3-8. Figure 3C-72 from EPA (2020b).

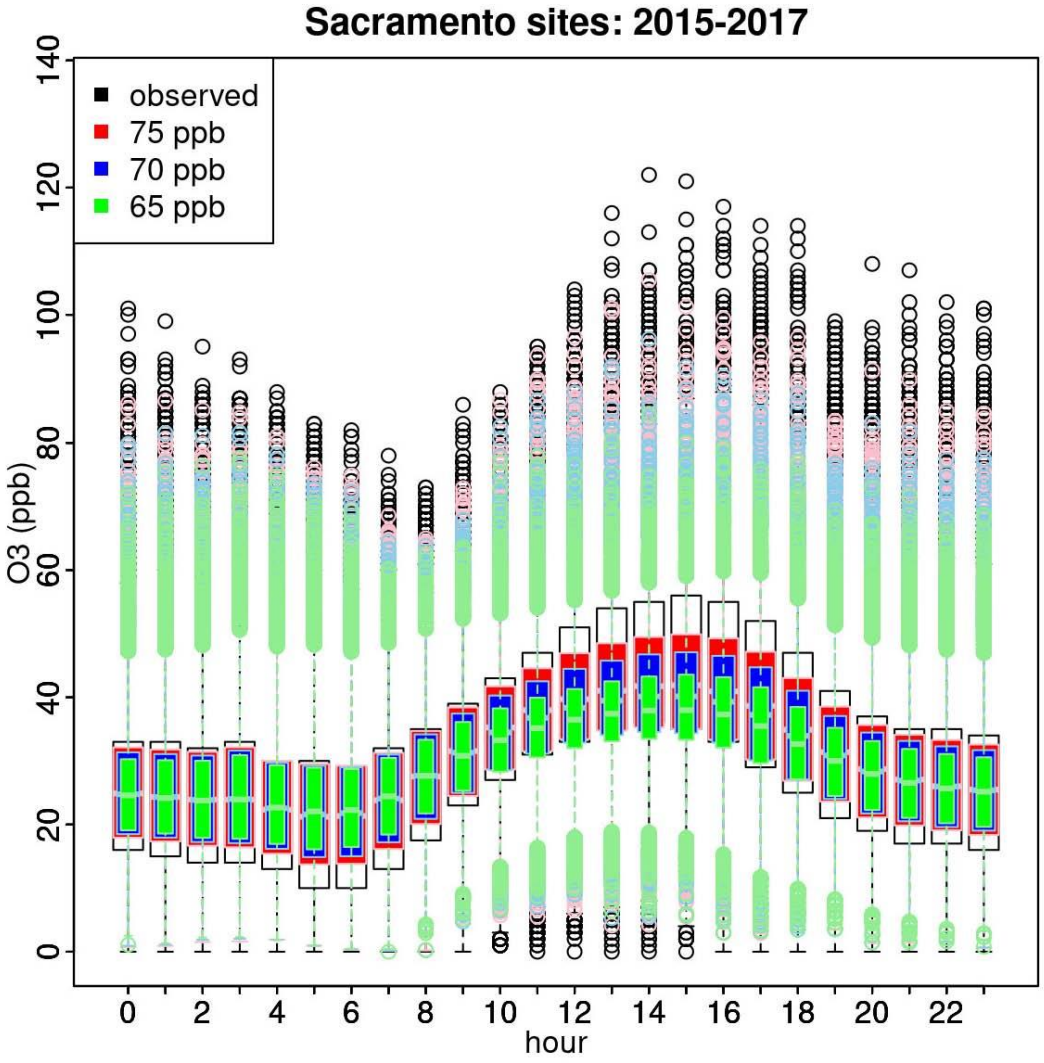


Figure 3C-73. Diurnal distribution of hourly O_3 concentrations at monitoring sites in Sacramento.

Figure 3-9. Figure 3C-73 from EPA (2020b).

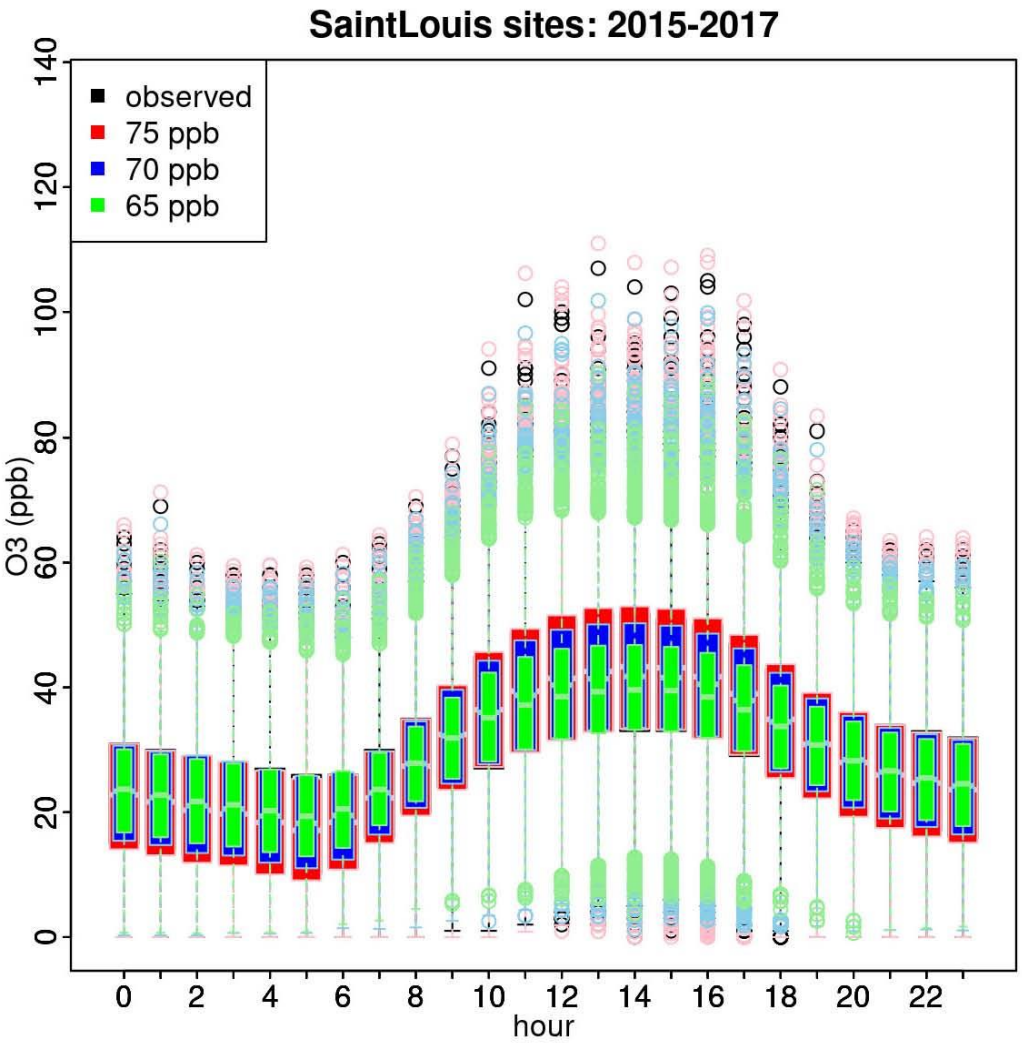


Figure 3C-74. Diurnal distribution of hourly O_3 concentrations at monitoring sites in St. Louis.

Figure 3-10. Figure 3C-74 from EPA (2020b).

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.

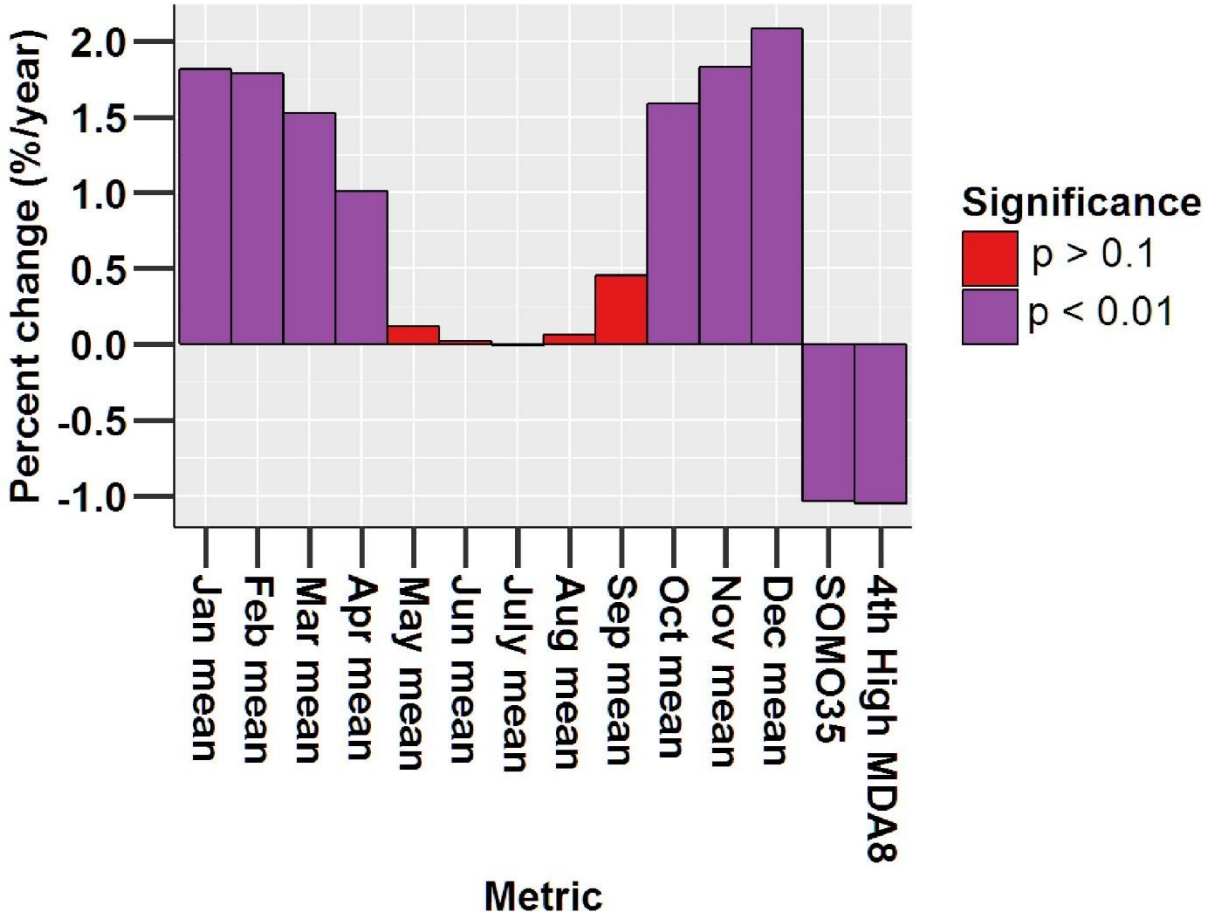


Figure 3-11. The Theil-Sen (%/year) trend in monthly average O_3 levels and the annual SOMO35 and 4th highest MDA8 human health metrics (A4MDA8) for a suburban site for 1980-2013 in Philadelphia, Pennsylvania (EPA AQS ID: 421010024-1). The $p < 0.05$ value was used to determine significance using the 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 U.S. 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, as anticipated, 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.

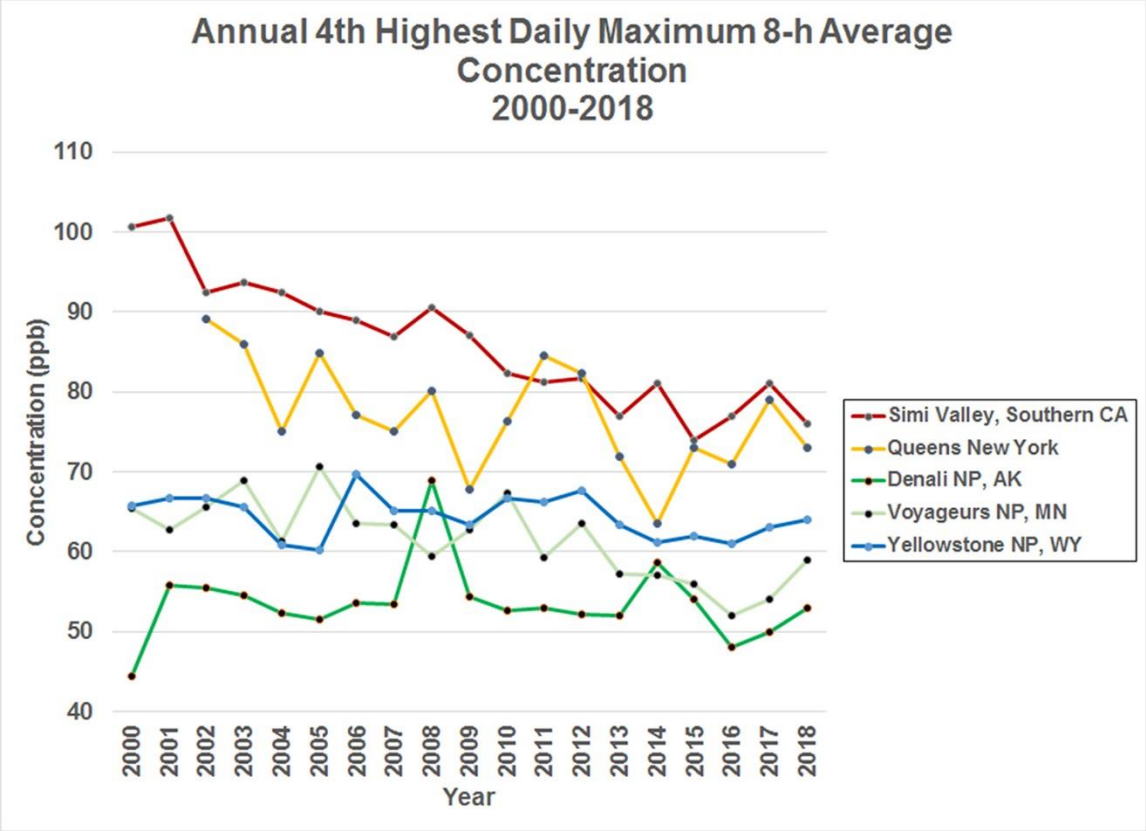


Figure 3-12. The annual 4th highest daily maximum 8-h average O_3 concentration for the period 2000-2018 for Simi Valley, CA (061112002), Queens New York, NY (360810124, Denali National Park, AK (020680003), Voyageurs National Park, MN (271370034), and Yellowstone National Park, WY (560391011).

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.

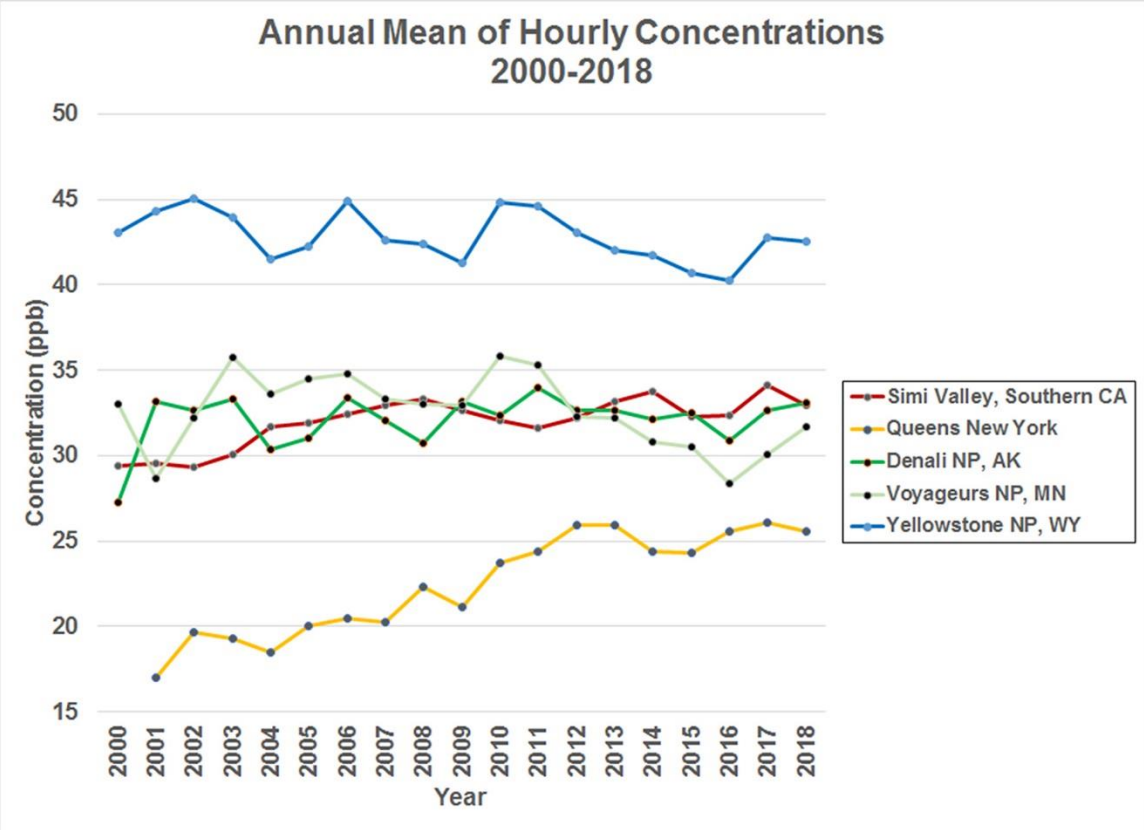


Figure 3-13. The annual mean of the hourly average O_3 concentrations for the period 2000-2018 for Simi Valley, CA (061112002), Queens New York, NY (360810124, Denali National Park, AK (020680003), Voyageurs National Park, MN (271370034), and Yellowstone National Park, WY (560391011).

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 annual average values at many of the O_3 monitoring sites have 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 annual 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 U.S. Based on the annual mean of hourly average concentrations, the high-elevation Yellowstone National Park site experiences much higher 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; EPA, 2014a). 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 daily maximum 8-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 daily maximum 8-h average O_3 concentrations 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 Fig. 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.

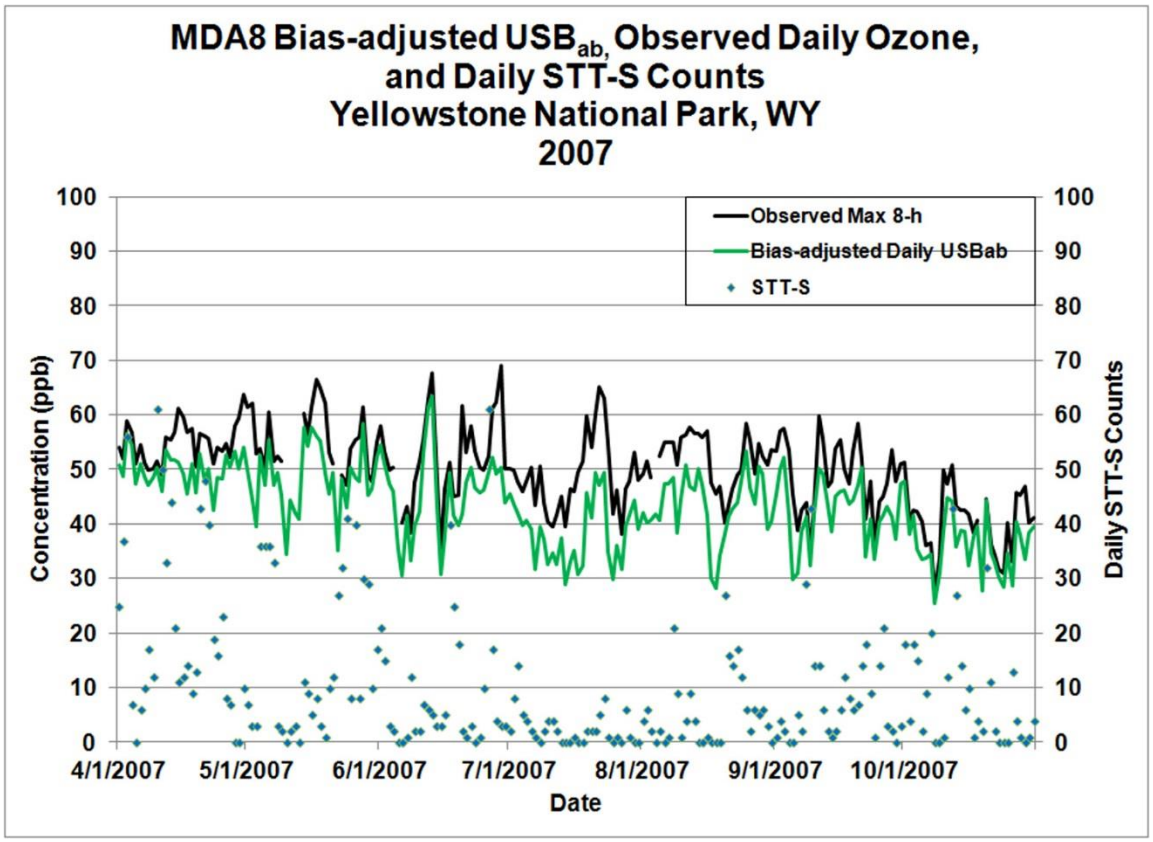


Figure 3-14. A comparison of the observed 8-h daily maximum concentration with the estimated bias-adjusted U.S. Background (USB_{AB}) 8-h daily maximum concentrations for Yellowstone National Park (AQS ID 560391011) for April-October 2007. The daily stratospheric-tropospheric transport to surface (STT-S) 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.

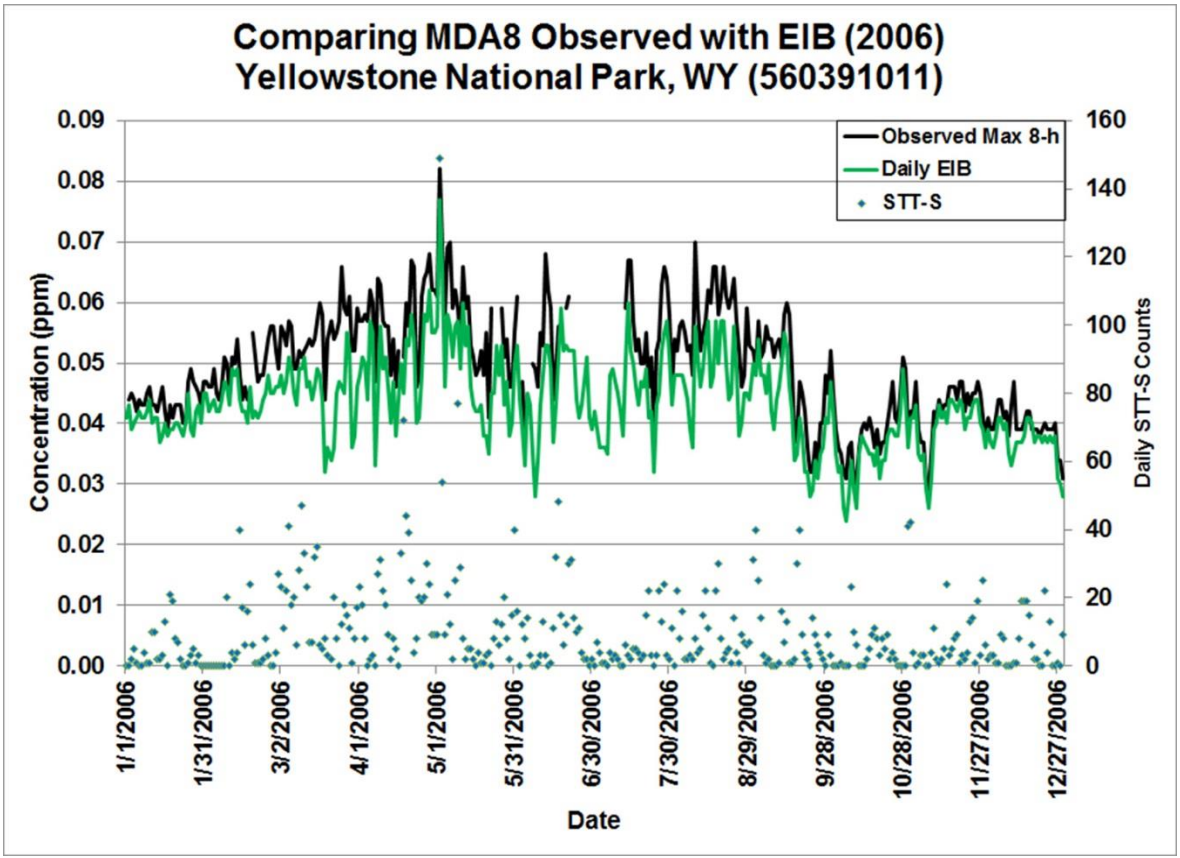


Figure 3-15. A comparison of the observed 8-h daily maximum concentration with the estimated bias-adjusted Emissions Influenced Background (EIB) 8-h daily maximum concentrations for Yellowstone National Park (AQS ID 560391011) for January-December 2006. The daily stratospheric-tropospheric transport 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 U.S. 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 show increasing trends using the annual average concentration metric.

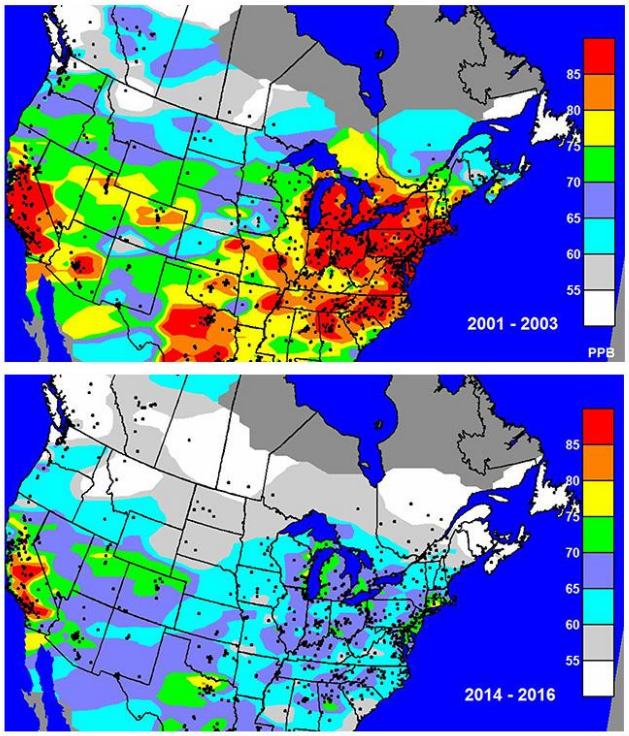
The calculation of the annual mean concentration includes the low, middle, and high hourly average concentrations. Lefohn et al. (1998), Simon et al. (2015) and Lefohn et al. (2017, 2018) have discussed the effects of NO_x emissions on hourly average O_3 concentrations. As pointed out in the PA (EPA, 2020b), 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. Ozone is titrated by NO, with the result that the frequency of low hourly average values increases, and the frequency of the higher hourly levels increases due to precursor 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) indicated 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. 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 pattern has been described in the literature (Lefohn et al., 2017, 2018). As noted in Section 1.8, the COVID-19 lockdowns that occurred in 2020 throughout the world during the spring (Norther Hemisphere) and fall (Southern Hemisphere) resulted in O_3 increases when averaging metrics were used and O_3 decreases when MDA8 metrics were applied.

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. 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 What Months the Highest O_3 Concentrations Occur as Emissions are Reduced

In the U.S., 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, Figs. 3-17 to 3-24 (Fig. 3C-75, page 3C-111) through Fig. 3C-82, page 3C-118) display the same information as Figs. 3-3 to 3-10 (Figure 3C-67 through Fig. 3C-74 in the PA) but for monthly rather than diurnal distributions. The figures below illustrate the modeling results presented in the PA document (EPA, 2020b) 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.

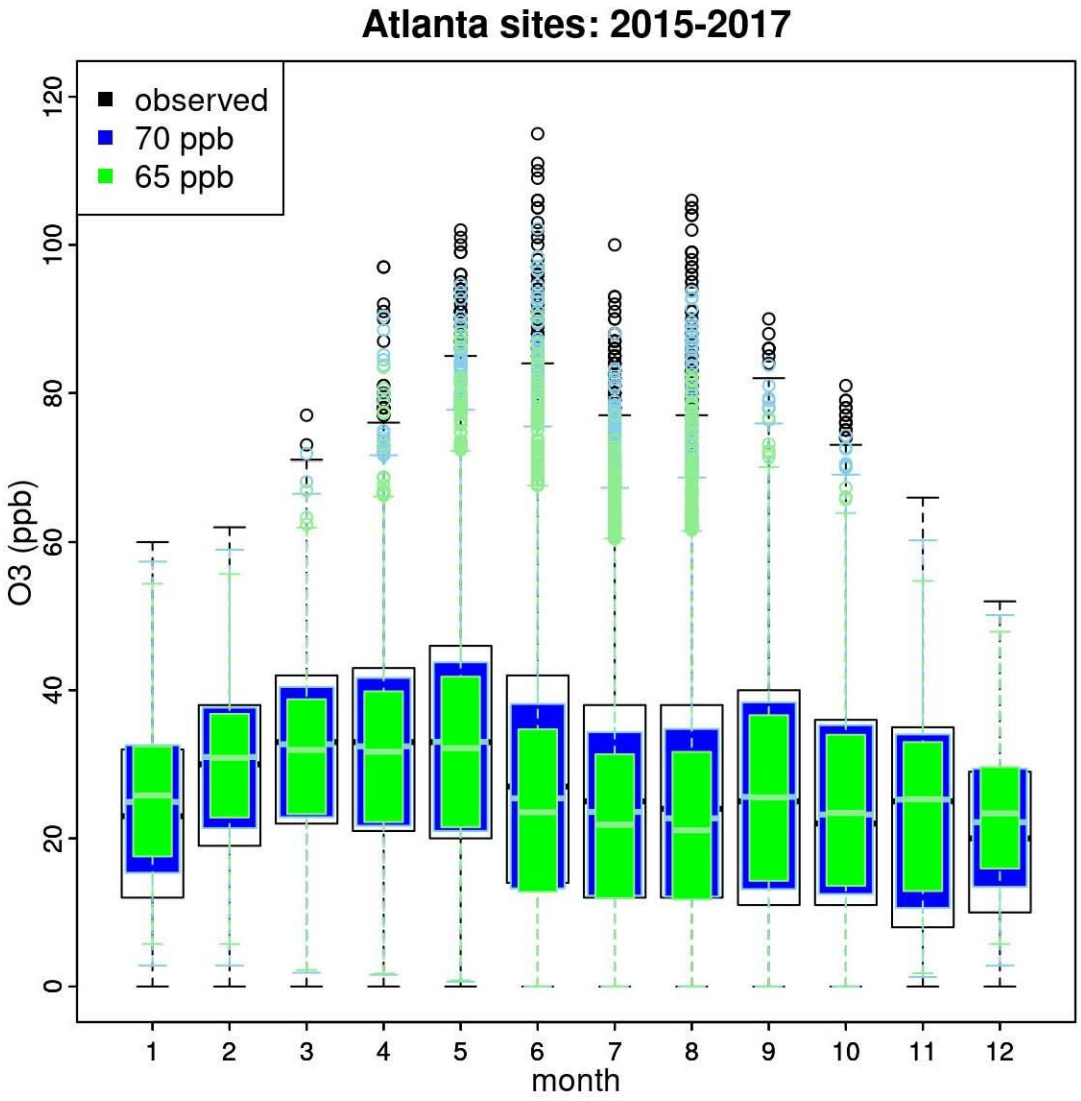


Figure 3C-75. Monthly distribution of hourly O_3 concentrations at monitoring sites in Atlanta.

Figure 3-17. Figure 3C-75 from EPA (2020b).

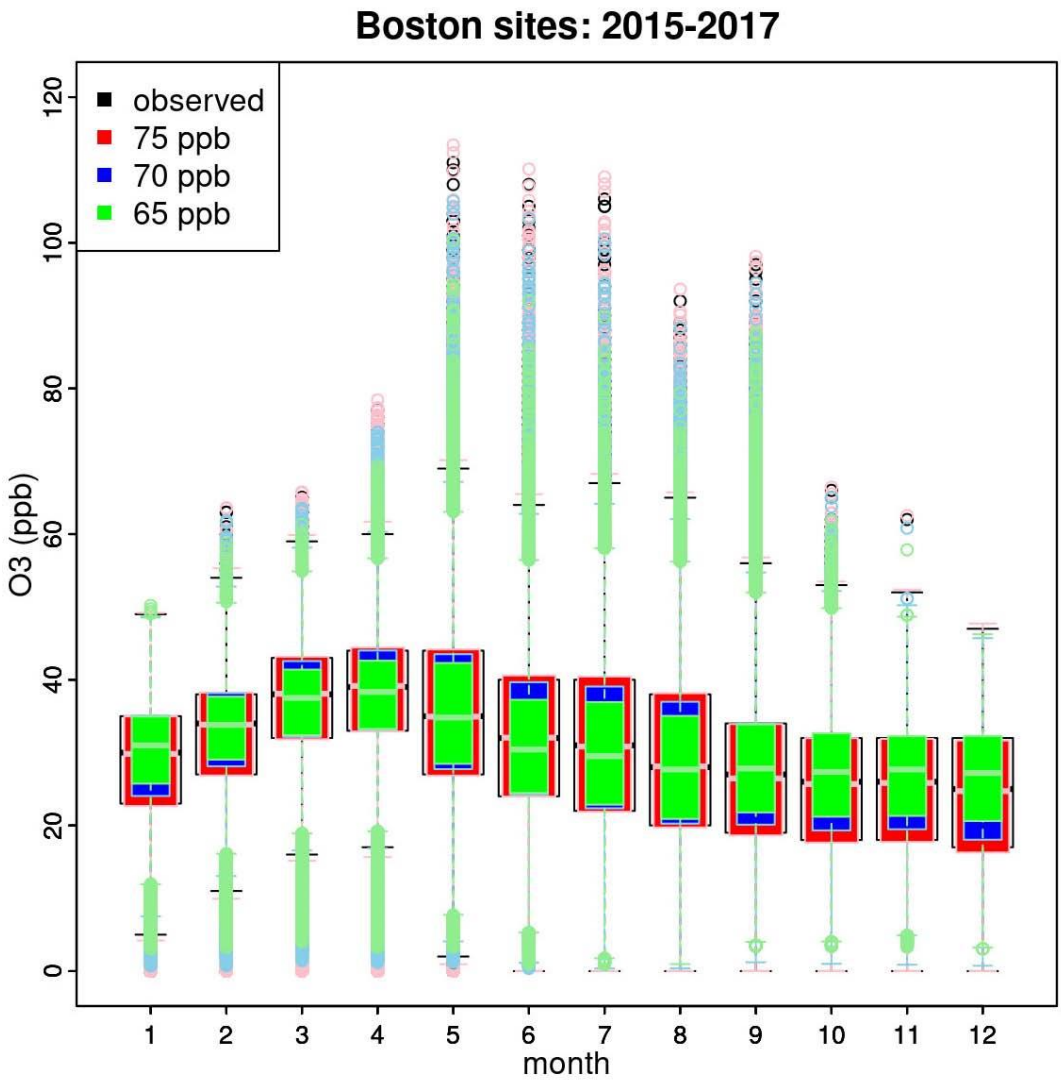


Figure 3C-76. Monthly distribution of hourly O_3 concentrations at monitoring sites in Boston.

Figure 3-18. Figure 3C-76 from EPA (2020b).

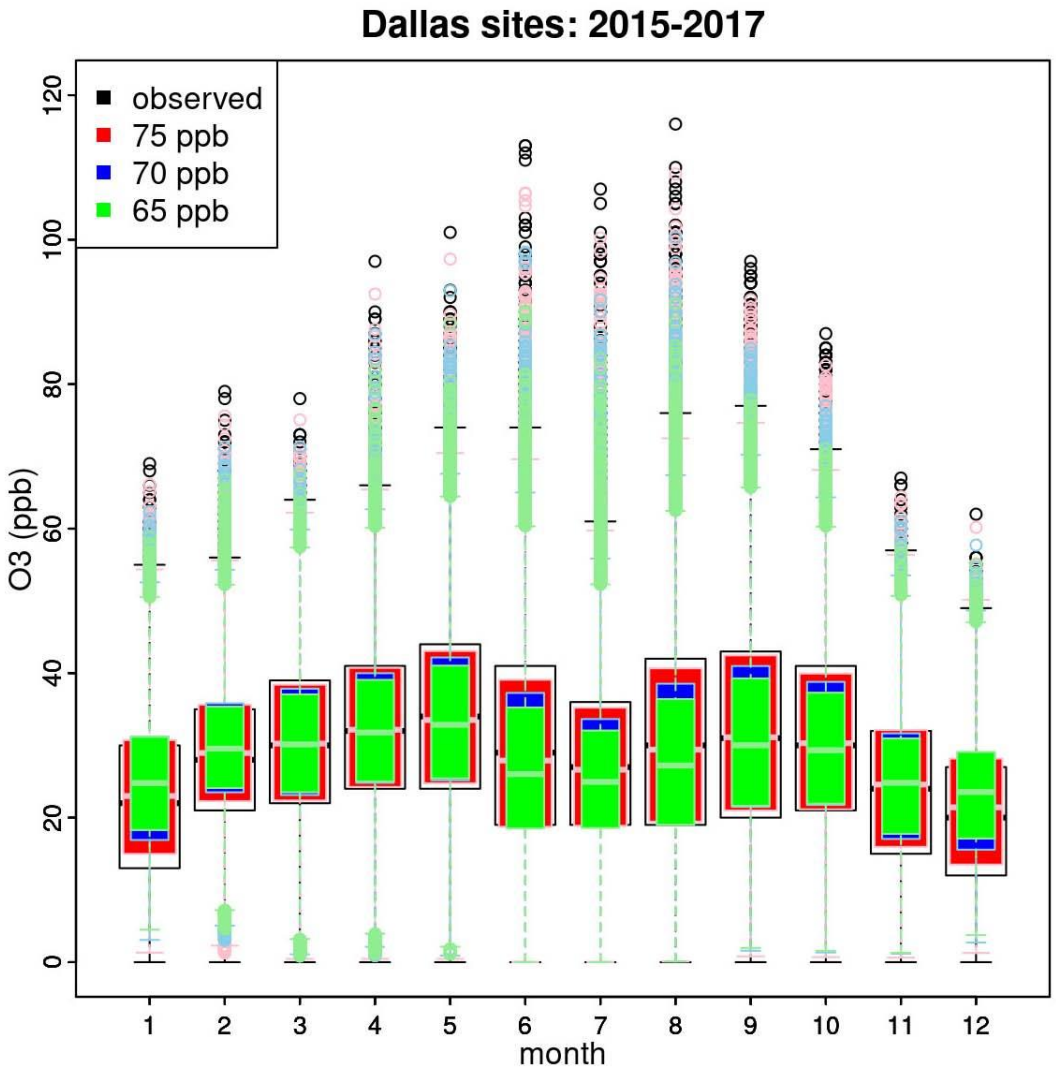


Figure 3C-77. Monthly distribution of hourly O_3 concentrations at monitoring sites in Dallas.

Figure 3-19. Figure 3C-77 from EPA (2020b).

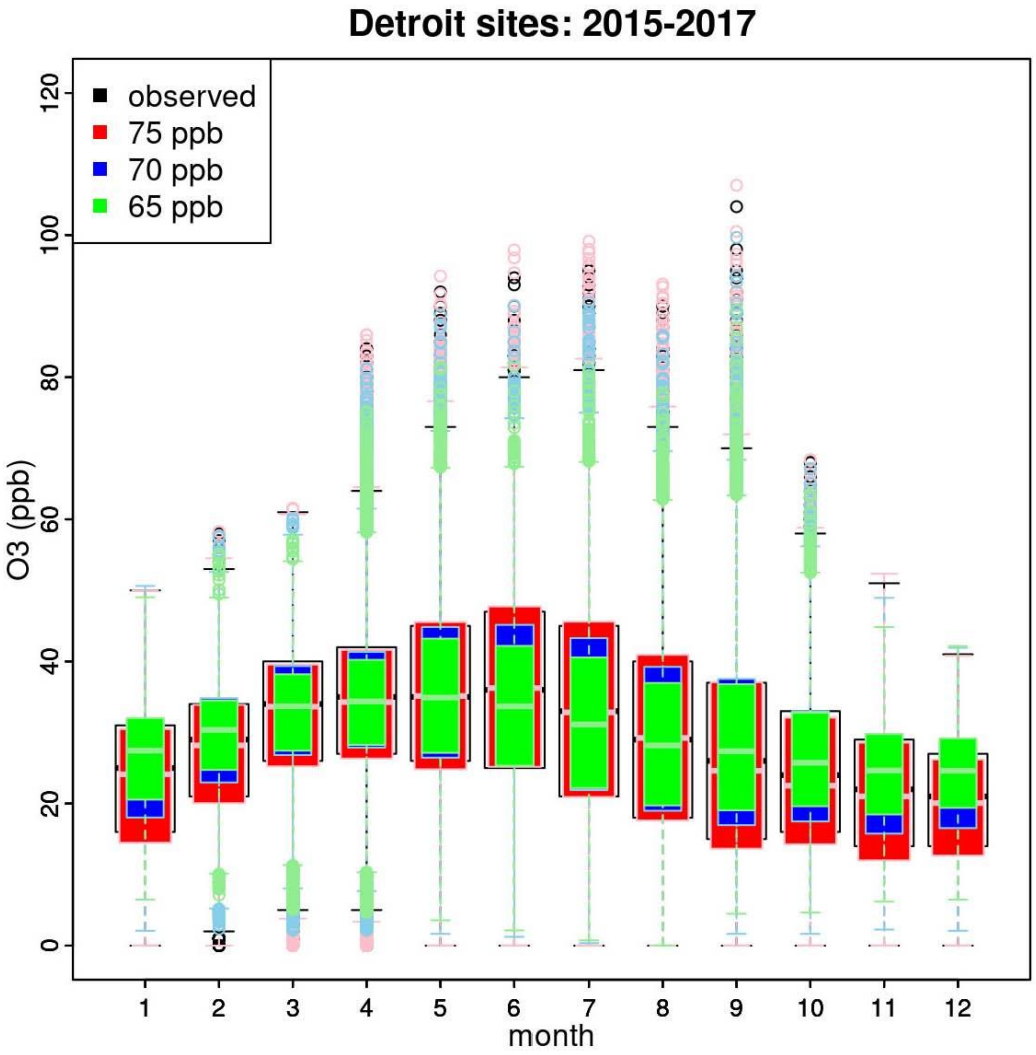


Figure 3C-78. Monthly distribution of hourly O_3 concentrations at monitoring sites in Detroit.

Figure 3-20. Figure 3C-78 from EPA (2020b).

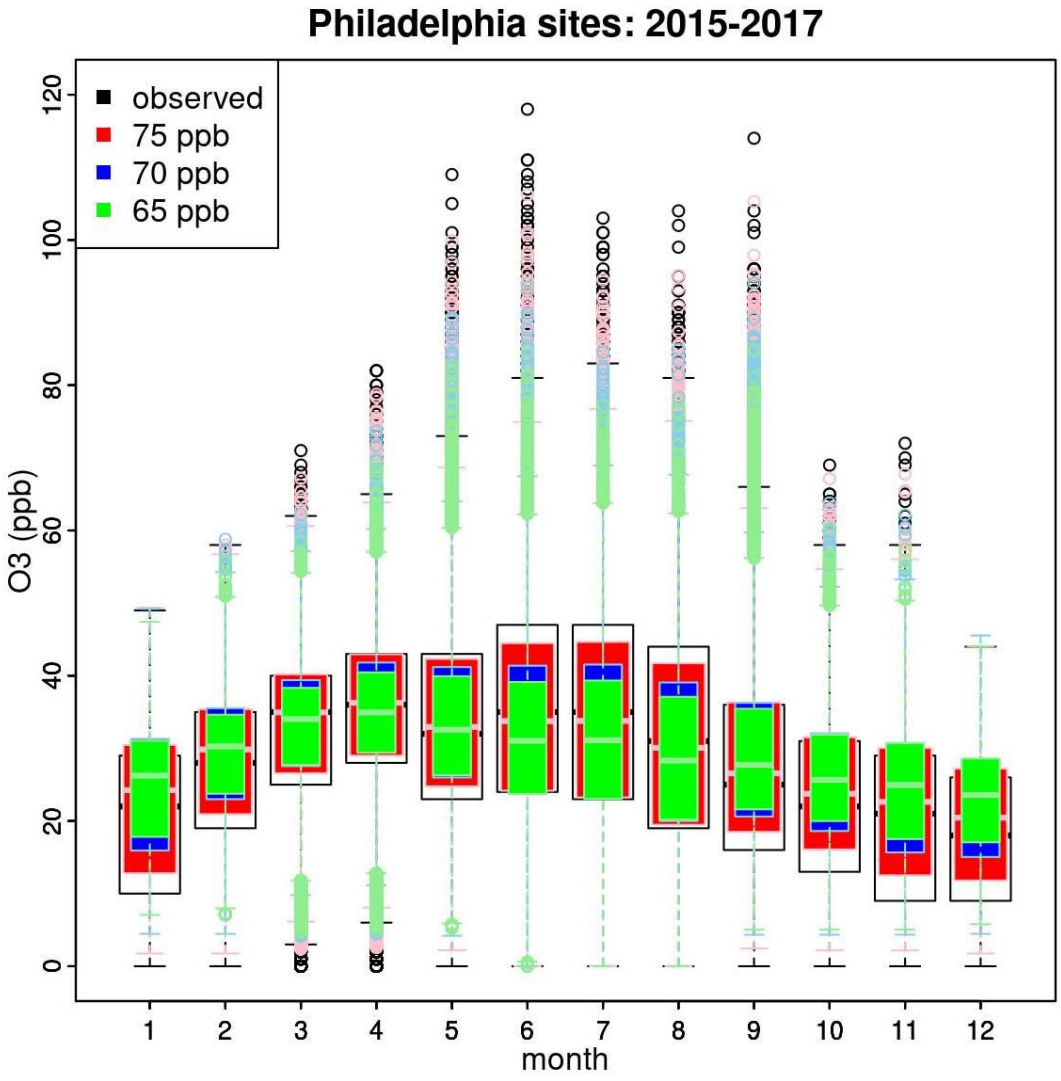


Figure 3C-79. Monthly distribution of hourly O_3 concentrations at monitoring sites in Philadelphia.

Figure 3-21. Figure 3C-79 from EPA (2020b).

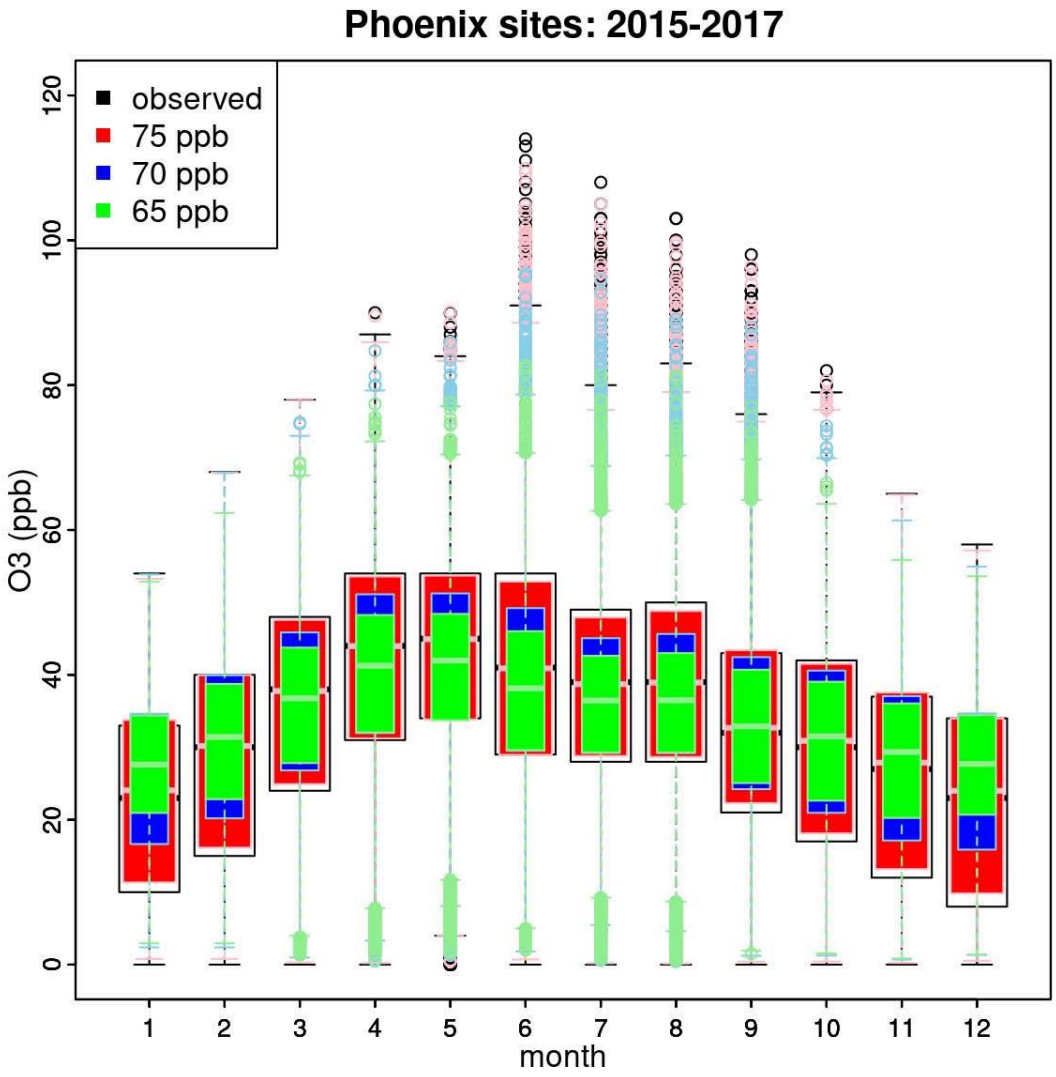


Figure 3C-80. Monthly distribution of hourly O_3 concentrations at monitoring sites in Phoenix.

Figure 3-22. Figure 3C-80 from EPA (2020b).

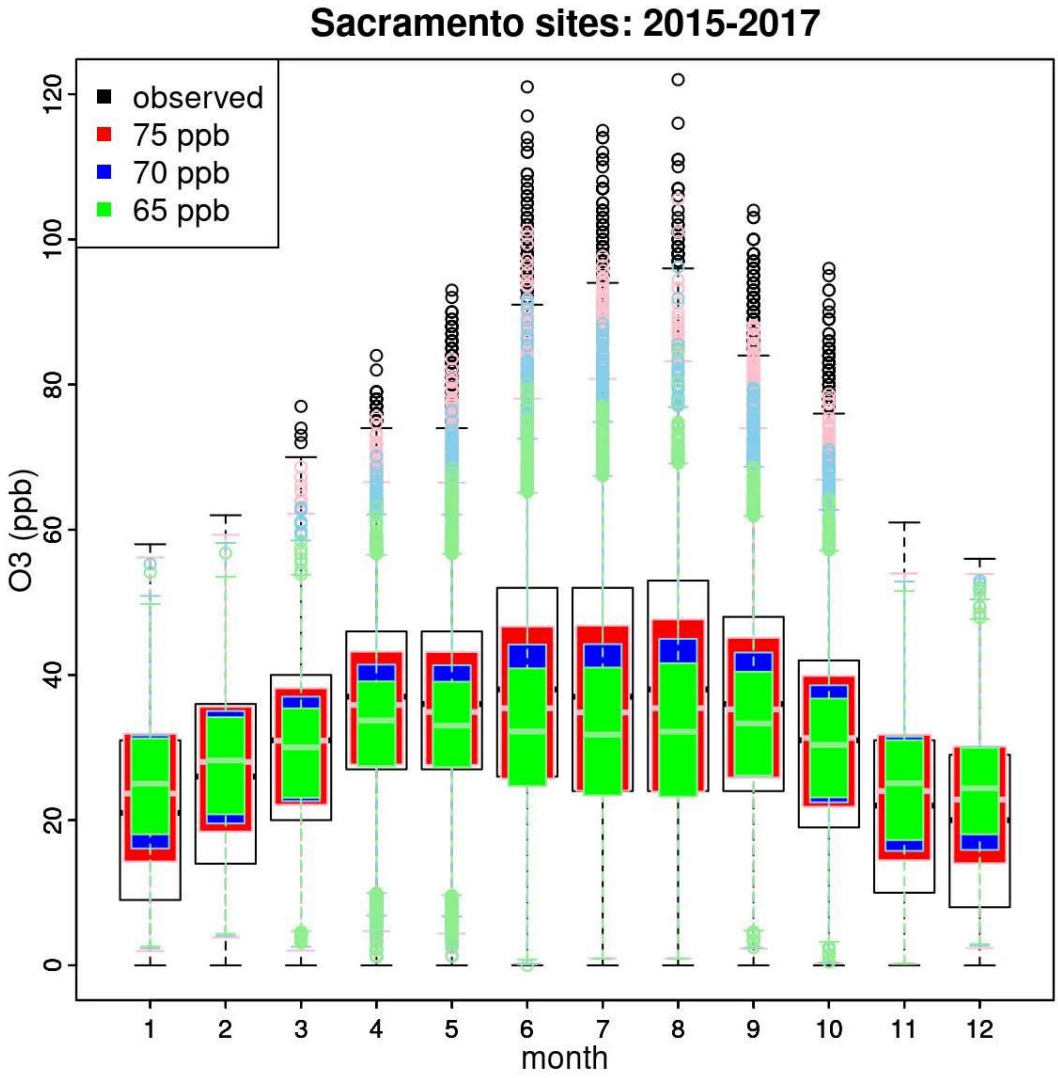


Figure 3C-81. Monthly distribution of hourly O_3 concentrations at monitoring sites in Sacramento.

Figure 3-23. Figure 3C-81 from EPA (2020b).

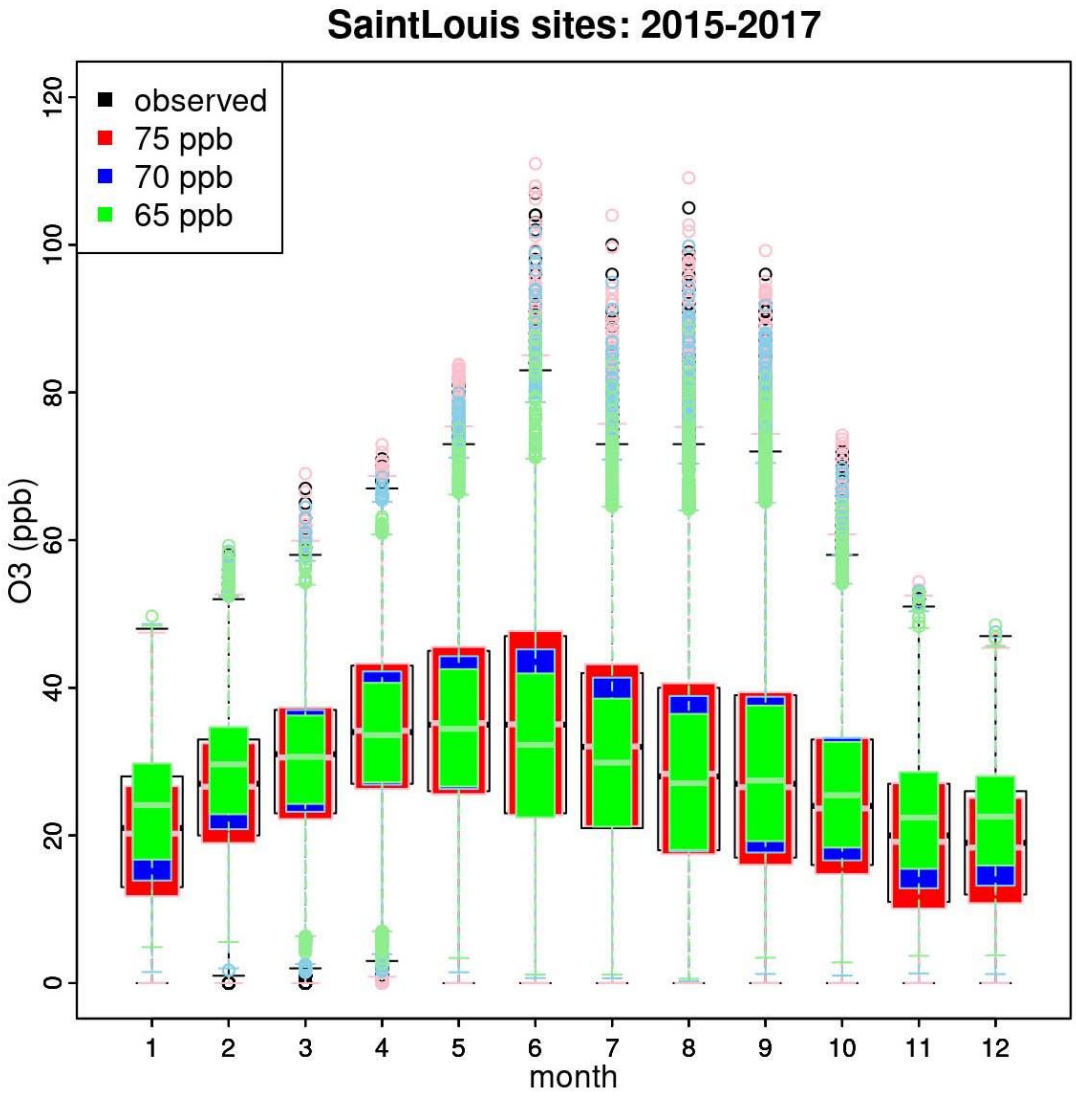


Figure 3C-82. Monthly distribution of hourly O_3 concentrations at monitoring sites in St. Louis.

Figure 3-24. Figure 3C-82 from EPA (2020b).

Using its models, for the monthly plots for the 8 cities, the EPA notes in the PA (EPA, 2020b) 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 (Fig. 3-20), Philadelphia (Fig. 3-21), Phoenix (Fig. 3-22), and St. Louis (Fig. 3-24). 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 intrusion and international transport, have been shown to peak during the spring months as discussed in the ISA (EPA, 2020a, page IS-15).

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 U.S. during the springtime (March to mid-June). Using hourly average O_3 data from 57 National Park Service 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 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 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 (the first 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 (≥ 50 ppb) at both high- and low-elevation monitoring sites across the U.S. 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 U.S., 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 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 PA (EPA, 2020b), 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 the April–June period. 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 U.S. 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 109 and 110, illustrate that while the highest O_3 exposures at the high-elevation Yellowstone National Park site may occur during the spring months, STT-S 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: 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: 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 ISA (EPA, 2020a) and the PA (EPA, 2020b) that the EPA is focused on answering the following question:

- How much of the current O_3 can be attributed to sources other than U.S. anthropogenic sources?

On page 2-27 in the PA, the authors note:

> In this review, as in past reviews, the EPA generally characterizes O_3 concentrations that would exist in the absence of U.S. anthropogenic emissions as U.S. background (USB). ***An alternative phrasing for USB is the O_3 concentrations created collectively from global natural sources and from anthropogenic sources existing outside of the U.S. 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 (ISA, section 1.2.2.*** (emphasis added).

A great deal of the focus by EPA in the modeling effort (page 1-52 of the ISA (EPA, 2020a)) appears to be on answering the above question as noted below:

- Applications of chemical transport models (CTMs) to estimate USB O_3 have found that USB concentrations are relatively constant with increasing total O_3 concentration, indicating that days with higher O_3 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 O_3 concentration as the most relevant for discussing USB O_3, 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 O_3 in contributing to the highest O_3 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 1-66 in the ISA (EPA, 2020a) 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 O_3 concentrations have been generally predicted to be similar to or smaller than seasonal mean USB O_3 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 O_3 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 O_3 concentration.

Further, in the PA (2020b), this focus continues on page 2-66 of the PA where the Agency provides reasons for its interest in background O_3 when it notes in its summary bullets:

- Predicted international contributions, in most places, are lowest during the season with the most frequent occurrence of MDA8 concentrations above 70 ppb. Except for the near-border areas, the International contribution requires long-distance transport that is most efficient in Spring.

- The USA contributions that drive predicted MDA8 total O_3 concentrations above 70 ppb are predicted to typically peak in summer. In this typical case, the predicted USB is overwhelmingly from Natural sources. The most notable exception to the typical case is reflected by predictions for an area near the Mexico border where the modeling indicates that a combination of Natural and Canada/Mexico contributions can lead to predicted MDA8 USB concentrations 60-80 ppb, on specific days, which is consistent with the previous O_3 PA (Section 2.4).

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 1-52 of the ISA (EPA, 2020a), 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. 3-26 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 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 (id. at 622-23). Thus, the court concluded that the EPA did not act unlawfully or arbitrarily or capriciously in setting the 2015 NAAQS without regard for background O_3 (id. at 624).

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. As noted in the PA (EPA, 2020b, page 2-52), when episodic natural events contribute to elevated O_3 concentrations documented in the air quality monitoring data to such an extent that they result in a regulatorily significant exceedance or violation of the NAAQS, the data can be addressed via the Exceptional Events Rule (40 CFR 50.14). Given the current level of the 8-h daily maximum concentration of 70 ppb for the O_3 NAAQS, the Exceptional Events Rule was applied by the states in several instances. For example, for the period 2017-2019, Arizona, California, Colorado, Idaho, Louisiana, Montana, Nevada, Utah, and Wyoming requested that specific hourly average O_3 concentrations reported for specific sites in their states be considered under the Exceptional Events Rule.

As noted in Section 1.5 earlier, while background O_3 currently is not a consideration in the setting of the level of the O_3 standard, background O_3 is an important consideration for assessing human health effects risks. The risk assessments play an important role in the margin of safety determinations. Background O_3 concentrations in the low- and mid-level part of the distribution of concentrations make up a large fraction of the total ambient O_3 levels and potentially can influence human health risk assessments associated with margin of safety determinations. An adequate margin of safety is a policy choice left specifically to the Administrator's judgment. The greater the contribution of background O_3 to the human health risk assessment, the greater the uncertainty will be to the input into the margin of safety consideration. Thus, currently because of its importance in affecting the human health risk assessments used in the margin of safety determination, background O_3 is an important consideration to be quantified for those 8 cities highlighted in the Agency's modeling analyses (i.e., Atlanta, Boston, Dallas, Detroit, Philadelphia, Phoenix, Sacramento, and St. Louis) in the PA (EPA, 2020b). Because background O_3 for the 8 cities was not performed, no information was provided in the current O_3 NAAQS rulemaking process concerning the contribution of background to the human health effects risks that provide valuable information on the margin of safety consideration.

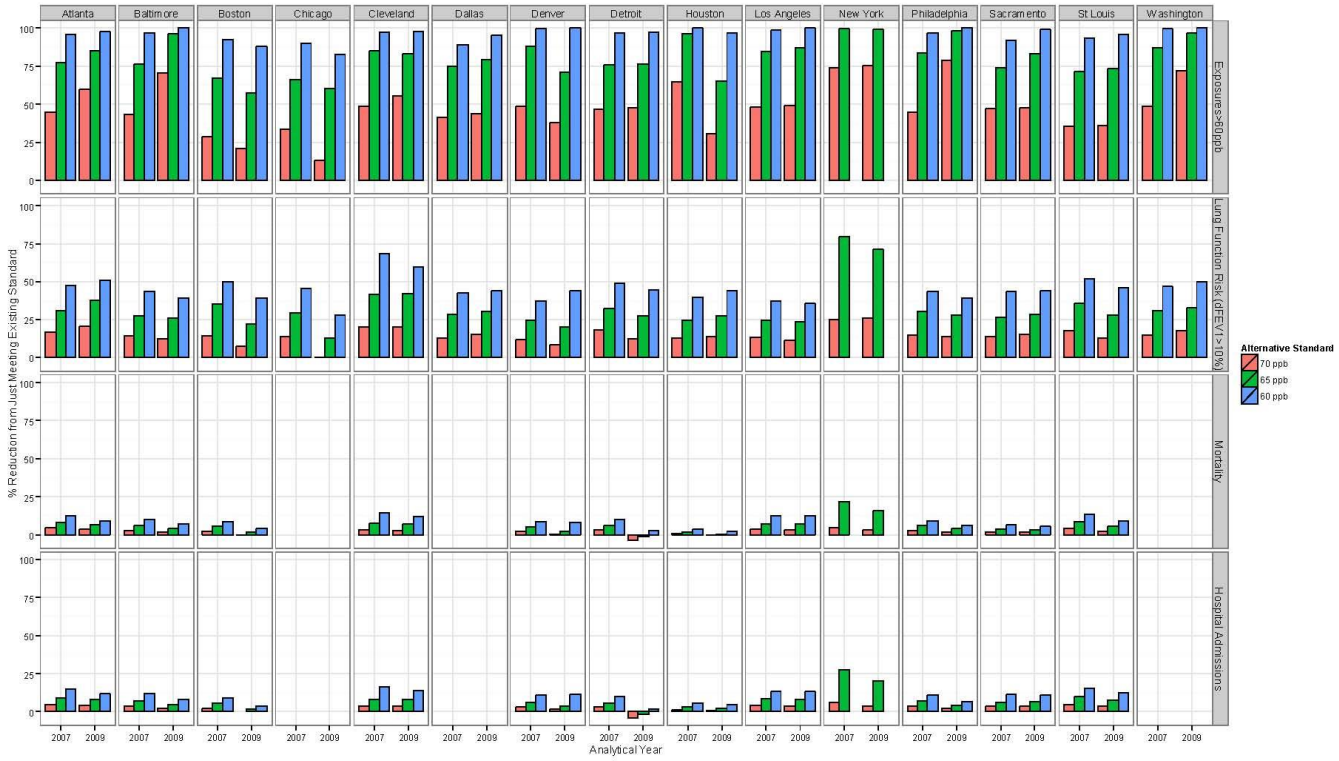


Figure 9-8. Comparison of the Percent Reduction in Key Risk Metrics for Alternative Standard Levels Relative to Just Meeting the Existing 75 ppb Standard.

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

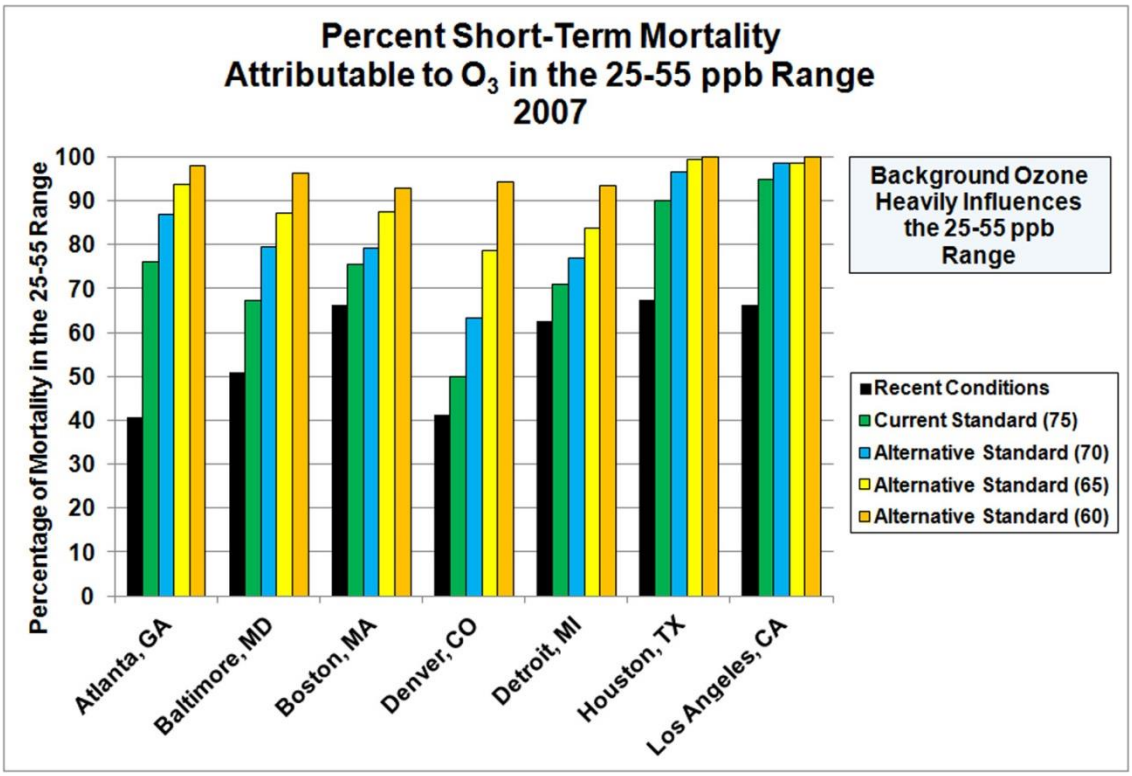


Figure 3-26. Percent short-term ozone-attributable mortality in the 25-55 ppb range for various exposure conditions for 2007 for 7 of 12 cities. (Source: Data from Fig. 7-B1 on page 7-B3 of EPA, 2014d).

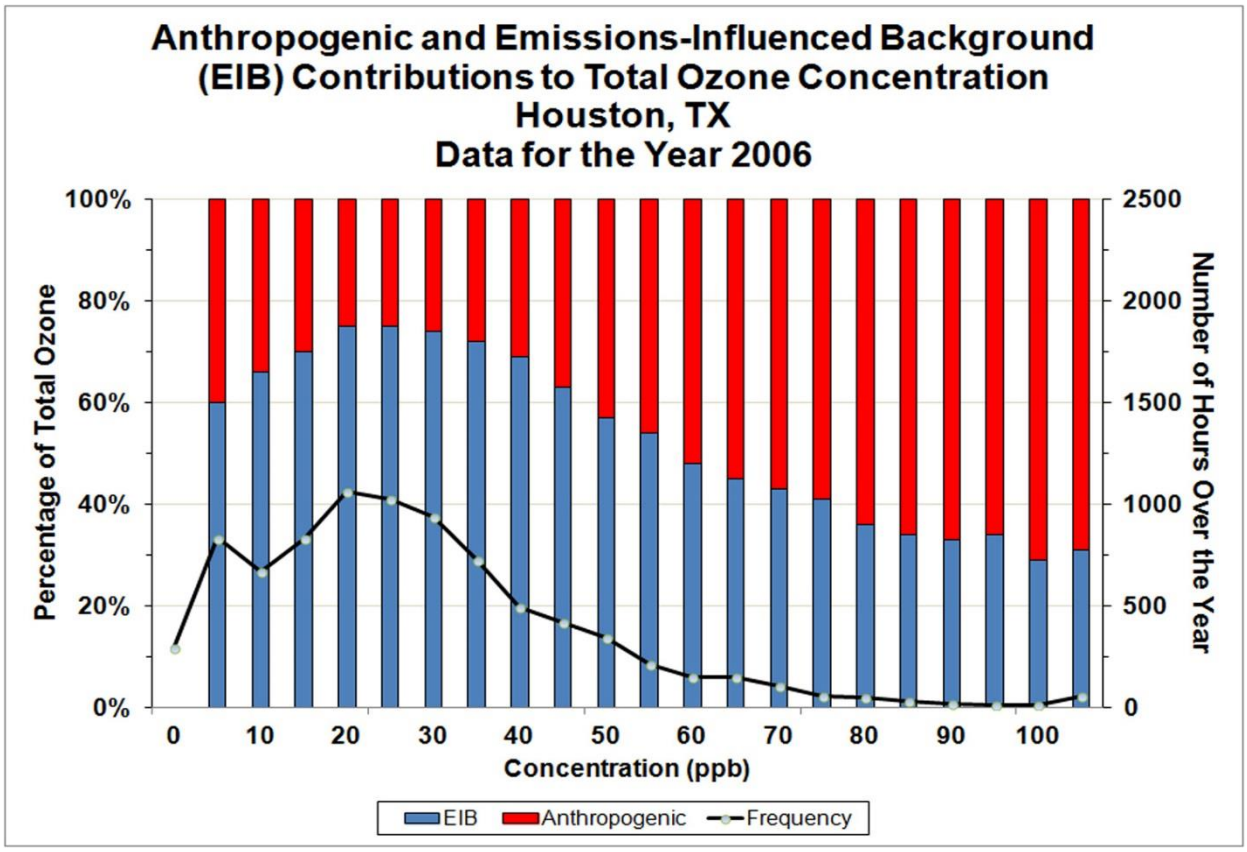


Figure 3-27. Binned (5 ppb) frequency distribution of observed hourly total O_3 (black curve; right axis) and average relative binned contributions of hourly maximum Emissions Influenced Background (EIB) and anthropogenic O_3 (bars; left axis) for Houston, TX (AQS ID 482010055) in 2006. Lefohn et al.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

3.2.2 How is the Term *Background O_3* Defined?

Based on the previous section, it appears that the EPA desires to answer the question **"How much of the current O_3 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 discussed previously, quantifying background O_3 is also important for assessing human health and ecological effects risks.

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., 2012; 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 U.S. 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., 2012; 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,b). While considerable discussion in the U.S. 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 EPA) to include contributions from global anthropogenic and natural sources in the absence of North American (i.e., U.S., 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 2013 ISA (EPA, 2013) modified its definition of background by introducing the terminology U.S. 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 U.S. anthropogenic emissions. In other words, the USB O_3 concentration is defined as the O_3 concentration that would occur ***if all U.S. anthropogenic O_3 precursor emissions were removed*** (EPA, 2020a, 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 U.S. anthropogenic emissions. In August 2014 in its PA, the EPA (2014a) described estimates of source-apportionment U.S. Background (USB_{AB}). The EPA (2014a) defined source-apportionment based U.S. 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 U.S. 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 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.

On page 1-6 of the ISA (EPA, 2020a), 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 O_3 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 not representative of contributions in nearby lower elevation locations.

The ISA (EPA, 2020a) 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 ISA (EPA, 2020a) 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 (EPA, 2020a), stratospheric intrusion events are an exception. The ISA (EPA, 2020a) 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 O_3.

In summary, the following terms have been used in the ISA (EPA, 2020a) to describe background O_3:

- USB is defined to include contributions from global anthropogenic and natural sources in the absence of U.S. 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 (EPA, 2013). NAB has also been referred to as policy-relevant background (PRB) in earlier publications (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 U.S. 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 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 ISA (EPA, 2020a) notes that this approach is consistent with the 2006 Ozone Air Quality Criteria Document (AQCD) (EPA, 2006a) and the 2013 Ozone ISA (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 (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 the USB methodology for estimating background O_3. No clear reason is provided why the authors of the ISA (EPA, 2020a) 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 ISA (EPA, 2020a) 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 2-38 of the PA (EPA, 2020b):

> 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). This analysis follows the zero-out approach for simplicity of interpretation and consistency with previous EPA analyses. In urban areas, this approach will estimate higher natural and USB contributions when NO_X titration is present. The estimate, therefore, is an estimate of what concentrations could be without U.S. anthropogenic emissions and not the fraction of observed O_3 that is USB.

On page 2-59 of the PA (EPA, 2020b), the authors state:

> The overall findings of this assessment are consistent with the 2014 O_3 PA, with the EPA's Background Ozone whitepaper (EPA, 2015), and with the peer reviewed literature (e.g., Jaffe et al., 2018).

In the EPA White Paper (EPA, 2015) that is referred to in the PA (EPA, 2020b), 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 (NO_x), volatile organic compounds (VOC), methane (CH4), and carbon monoxide (CO). This definition of background is specifically referred to as U.S. background (USB).

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." EPA provided no clear rationale for this decision.

On page 1-5 of the ISA (EPA, 2020a), 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 (e.g., "zero-out" modeling). Apportionment-based USB (USB_{AB}) has been defined as the amount of O_3 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 and used both USB and USB_{AB}.

The ISA (EPA, 2020a) notes on page 1-56:

> 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 made the decision 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 (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 EPA in its 2014 PA (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 2014 PA (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 ISA (EPA, 2020a) (Page IS-16):

> 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 ISA (EPA, 2020a, page 1-56) 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 O_3*** (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 O_3 levels. Source: Source: 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 ISA (EPA, 2020a, 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 O_3 concentrations that could be achieved if U.S. anthropogenic sources were eliminated, USB_{AB} is an estimate of how much O_3 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 U.S. 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 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*. This is an important concept. By estimating USB_{AB} concentrations, one could obtain a lower estimate of background O_3 and quantify the percentage contribution of background O_3 to the total concentration. The percentage would provide important information to the EPA Administrator about the influence of background levels to those human health risk estimates that affect margin of safety determinations. A quantitative understanding of background O_3 is essential for air quality management. This is especially true given the lowering of the NAAQS O_3 levels and the associated increasing relative importance of background O_3 as domestic precursor emissions decrease. Using the information provided in Table 3-1 (Table 2-1 in the 2014 PA), it appears that USB_{AB} might have been a better modeling methodology to apply rather than USB to answer the question "How much of the current O_3 can be attributed to sources other than U.S. anthropogenic sources?" The USB_{AB} estimate would have provided a lower estimate of background O_3 than could be used to bracket the range of concentrations.

Thus, we are left with the dilemma of why the EPA estimated USB rather than USB_{AB} for estimating 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 2015 NAAQS O_3 rulemaking, the 2014 PA (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 U.S. both the USB and USB_{AB} findings contained within the 2014 PA (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 have provided less than optimum estimates than would have been obtained if the USB_{AB} methodology had been applied.

3.2.4 EPA's Conclusion on Background O_3 Modeling Estimates

EPA ISA (2020a, page 1-65) notes that while the seasonal mean USB concentration patterns may be important for identifying atmospheric processes leading to high USB concentrations and for understanding total O_3 exposures over long periods, ***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 ISA (EPA, 2020a), 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 1-65 in the ISA (EPA, 2020a), the EPA notes that the 2013 Ozone ISA (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 ISA (EPA, 2020a, page 1-65) believes the disagreement among researchers 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 ISA (EPA, 2020a, page 1-65) 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 O_3 concentrations at elevations >1 km in the western U.S., while below 1-km baseline O_3 concentrations were higher in spring.

EPA in its ISA (EPA, 2020a) did not resolve the conflicting conclusions about when seasonal mean background O_3 is greatest. EPA should have addressed the inconsistencies in its own latest modeling results concerning the seasonal patterns to assess the adequacy of the latest model predictions. The ISA (EPA, 2020a) dismissed 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) illustrated the vertical profiles of O_3 at 4 sites in the West (Trinidad Head, Cheeka Peak, Mt. Bachelor Observatory, and Chews Ridge). The authors noted that at low elevations at the four sites, mean spring O_3 levels were about 10 ppb higher than summer values, whereas above 1 km, median spring and summer values were comparable with summer.

Guo et al. (2018) note that their model may have exaggerated the relative importance of enhanced background O_3 resulting from soil NO_x 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.

In the PA (EPA, 2020b), the authors summarize their conclusions from the EPA's updated background O_3 modeling results for the year 2016. These conclusions are as follows:

- This analysis focused on characterizing USB from Natural, International and USA contributions. For the analysis of the International component, the contributions from India, China, and international shipping peak during the spring when MDA8 O_3 is typically low. (page 2-59).

- For this analysis we did not attempt to quantify the contributions from individual Natural sources (e.g., lightning, soil, fires, stratosphere) or to address exceptional events beyond basic screening to remove very large fire plumes. (page 2-66).

- The USA contributions that drive predicted MDA8 total O_3 concentrations above 70 ppb are predicted to typically peak in summer. In this typical case, the predicted USB is overwhelmingly from Natural sources. The most notable exception to the typical case is reflected by predictions for an area near the Mexico border where the modeling indicates that a combination of Natural and Canada/Mexico contributions can lead to predicted MDA8 USB concentrations 60-80 ppb, on specific days, which is consistent with the previous O_3 PA (Section 2.4). (page 2-66)

- Consistent with previous assessments, USB is higher in the West than in the East. (page 2-65).

- Days for which MDA8 total O_3 concentrations are predicted to be above 70 ppb tend to have a substantially higher model-predicted USA (anthropogenic) contribution than other days in both the West and the East. (page 2-67).

- Predicted international contributions, in most places, are lowest during the season with the most frequent occurrence of MDA8 concentrations above 70 ppb. Except for the near-border areas, the International contribution requires long-distance transport that is most efficient in Spring. (page 2-67).

- The West has higher predicted USB concentrations than the East, which includes higher contributions from International and Natural sources. Within the West, high-elevation and near-border areas stand out as having particularly high USB. The high-elevation areas have more International and Natural contributions than low-interior areas in the same region. The near-border areas in the West can have substantially more international contribution than other parts of the West. (page 2-66).

- The current analysis indicates that natural and USA O_3 contributions peak during the traditional O_3 season (May through September), while long-range intercontinental transport of international O_3 (i.e. contributions from China, India etc.) peaks in the spring (February through May). (page 2-64).

- The natural contribution has a single maximum in late summer in the West, whereas, in the East there is evidence of two peaks— the largest in late Spring and a second peak in early Fall. (page 2-48).

- The Natural component of USB exhibits the largest magnitude difference between the West and East. (page 2-65).

- The contributions from Canada/Mexico at near-border locations are associated with relatively short-range transport and the seasonality peaks during May through September, similar to USA anthropogenic O_3. (page 2-64).

- Ozone formed from anthropogenic emissions in Canada and Mexico can peak in late spring or early summer when total O_3 is high. (page 2-65).

- Long-range transport and USA anthropogenic contributions tend peak at different times of the year, so the contribution of international is often at its minimum when local sources are the driving factor for high total O_3 during the May through September O_3 season. (page 2-65).

- On high O_3 days (greater than 70 ppb) the West-East differences are largely associated with international contributions in near-border areas and natural contributions at high-elevation locations. (page 2-65).

- In the northern hemisphere, the natural NO_x sources with the largest emissions are lightning (9.4 megatonN/yr), soils (5.5 megatonN/yr), and wildland fires (~2.2 megatonN/yr). (page 2-66).

- Because NO_x is the limiting precursor at hemispheric scales, the emissions data suggests that lightning and soils contribute are most likely the largest contributors to Natural O_3. As noted by Lapina et al. (2014), a large contribution from lightning may be the result of lightning strikes outside the U.S. while the contribution from soil NO_x tends to largest from emissions within the U.S. The distant lightning source is likely to have its effect as part of the well-mixed background. The local soil NO_x emissions have a clear seasonal cycle and is known to have large local contributions. (page 2-66).

- Wintertime O_3 events can be associated with emissions from local oil and gas production in the Intermountain West. Even though these episodes can occur as early in the year as February, international emissions may not contribute to them substantially. The conditions associated with these events result in decoupling of the local air masses from the upper atmosphere, essentially isolating air in the mountain valleys from the atmosphere above and reducing the influence of long-range transport compared to other winter and early spring days. As a result, these unique wintertime O_3 episodes may have little relative influence from international emissions despite occurring at a time of year when long-range transport from Asia is efficient. (page 2-65).

In the PA (EPA, 2020b), the EPA provides summary figures that are output from the 2016 USB analysis. For Fig. 3-28 (reproduced from Fig. 2-23 in the PA), the authors note on page 2-48 of the PA the following:

> The temporal pattern in the regional average clearly shows that the seasonality of MDA8 predictions for each total O_3 component varies by region. The natural contribution has a single maximum in late summer in the West, whereas, in the East there is evidence of two peaks— the largest in late Spring and a second peak in early Fall. The somewhat lower MDA8 O_3 in summer in the East requires further analysis but may be related to the lack of lightning emissions within the regional domain. The seasonality international contribution predictions is more similar between the two regions. The international contributions in both the West and East are greatest in Spring, but the contribution in the West is larger both at its peak and its trough, compared to the East. The total international contribution and the separately analyzed long-distance components (e.g., China, India, international shipping) peak in spring when O_3 lifetimes favor long-range transport (see Appendix 2B, Figure 2B-29). However, the Canada/Mexico component of international contributions peaks in summer because of the relative proximity to the U.S. receptors. The predicted USA contribution increases in the summer for both the West and the East, but the USA contribution in the West is smaller than in the East. As mentioned previously, this "all cells" average is disproportionately rural in the West. The following analysis looks further at the different types of land in the West, including urban areas that are more representative of population centers that behave differently than the "all cells" analysis.

Figure 3-28. Annual time series of regional average MDA8 O_3 concentration and 8-h contributions for the West (top) and the East (bottom). See legend above from the original figure. Source EPA (2020b Fig. 2-23, page 2-49).

On page 2-50 of the PA, the authors note that Fig. 2-24 (page 2-51 in the PA) illustrates the contributions to the West split into three parts: the highest elevation areas, the near border areas, and Low/Interior areas with a weighted average focusing on urban areas. Fig. 3-29 is from the PA (EPA, 2020b). The authors note the following:

> The urban area weighted average gives a larger weight to data in those urban areas that have dense emission sources (e.g., mobile). The urban area weighted average shows higher contribution from USA while Natural and International are lower compared to Figure 2-23. The differences between urban-weighted and non-weighted contributions are smaller in the East (not shown) than in the West (compare Figure 2-23 top and Figure 2-24 bottom). Compared to the West, the East has a larger fraction of land use that is urban (see Figure 2-22), which explains this difference. Thus, the non-weighted regional average contributions in the East includes the effects of urban areas much more so than the West. The seasonality of International is also different between the highest elevation areas, near border areas, and urbanized areas. At low/interior and at high-elevation sites, the simulated International contribution peaks earlier in the year than

at border sites. This earlier season peak is consistent with seasonality of O_3 lifetime necessary for long-range transport and a smaller contribution of long-distance sources (India, China, and Ships, see Appendix 2B, Figure 2B-30). At near-border sites, the seasonal cycle of predicted USB contributions from Canada/Mexico and from long-range transport combine to create a maximum later in the spring or early summer that is dominated by Canada/Mexico contributions (see Appendix 2B, Figure 2B-30, middle panel).

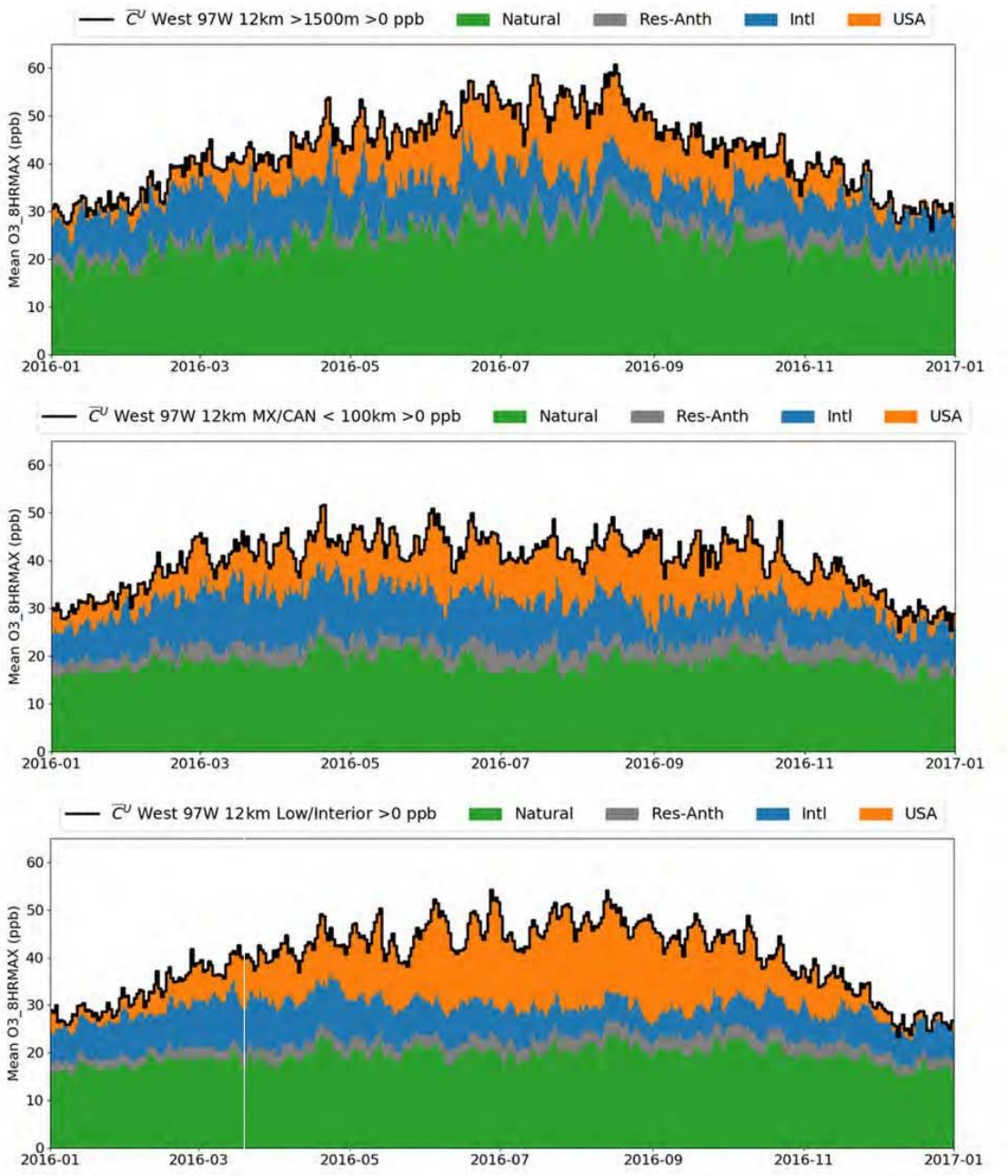


Figure 3-29. Annual time series of regional urban area-weighted average MDA8 O_3 concentration and 8-h contributions for the High-elevation West (top), near-boarder West (middle), and Low/Interior West (bottom). See legend above from the original figure. Source EPA (2020b page 2-51).

Earlier we had discussed specific applications of background O_3 in the standard-setting process. While seasonal mean background O_3 estimates are of interest for identifying atmospheric processes, their 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 ISA (EPA, 2020a) and PA (EPA, 2020b) 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 of equal importance for quantifying background O_3. In many cases, assessment of human health and ecological risks 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. Background O_3 affects the overall human health risk assessments, and therefore, can influences the margin of safety determinations required for establishing the O_3 NAAQS.

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 Fig. 3-30 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 in its 2014 PA (EPA, 2014a). In comparison, Fig. 3-31 illustrates that for Denver the contribution of background within the mid-range concentrations is approximately 75 to 80%. For the Los Angeles area (Fig. 3-32), a site heavily influenced by anthropogenic emissions, background contributes 60-80% in the mid-range.

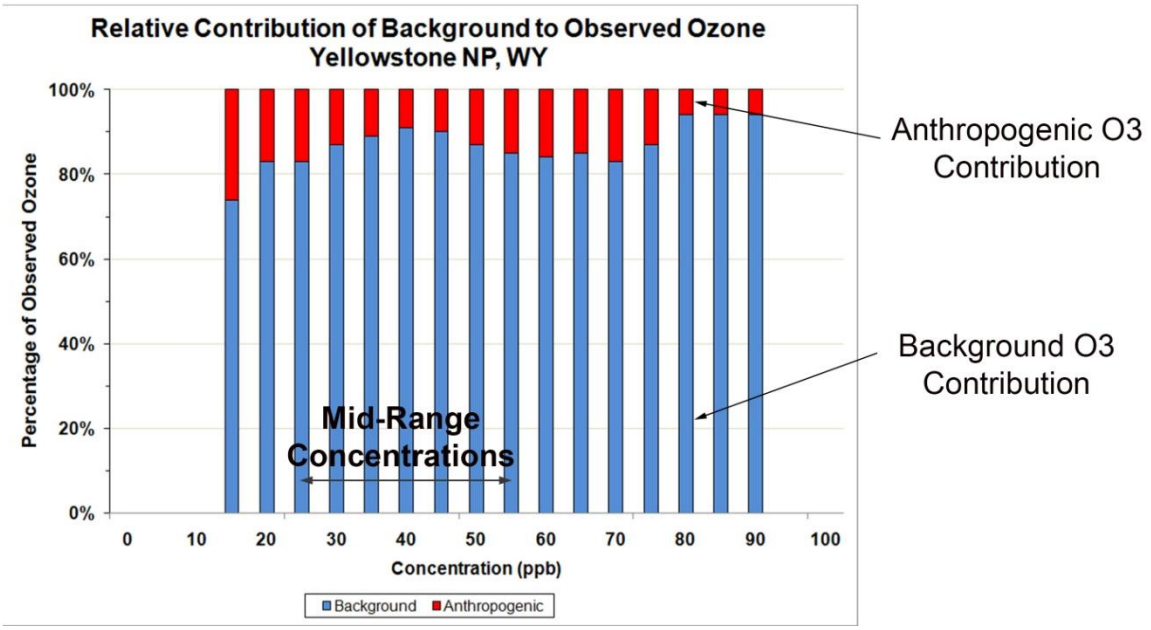


Figure 3-30. Average relative contributions of current hourly background (blue) and anthropogenic O_3 (red) for Yellowstone NP (WY) (AQS ID 560391011) in 2006. (Source: Lefohn et al., 2014).

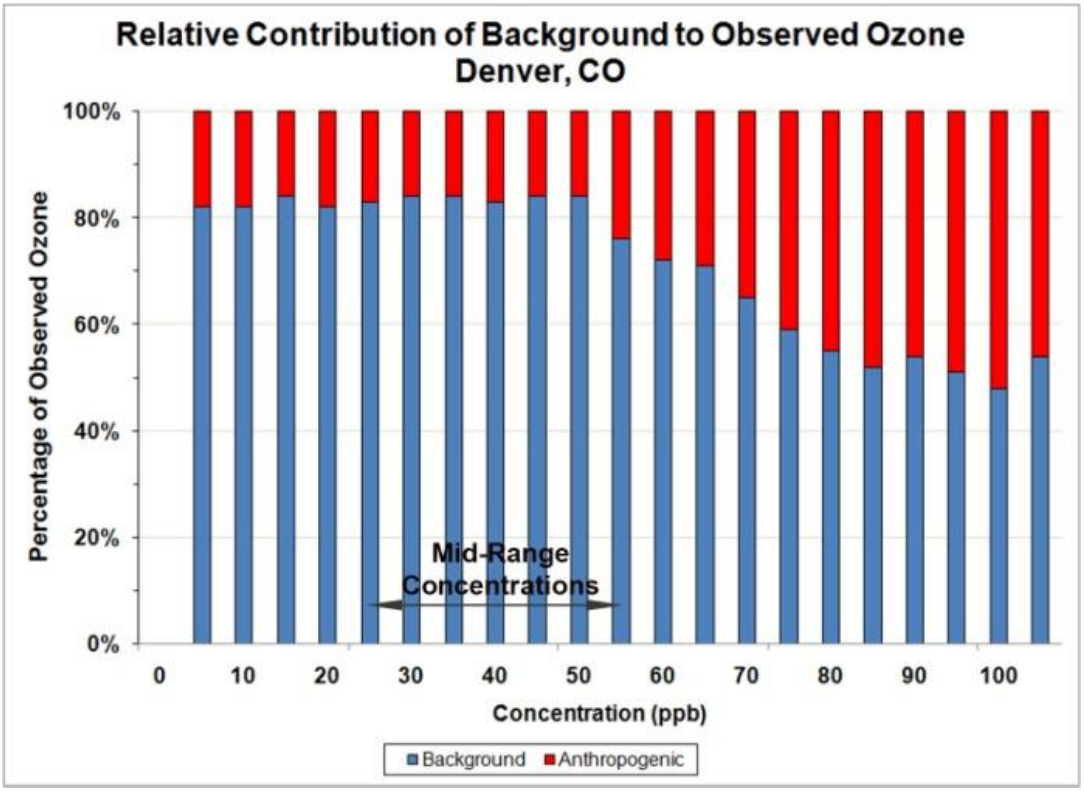


Figure 3-31. Average relative contributions of current hourly background (blue) and anthropogenic O_3 (red) for Denver (CO) (AQS ID 080590006) in 2006. (Source: Lefohn et al., 2014).

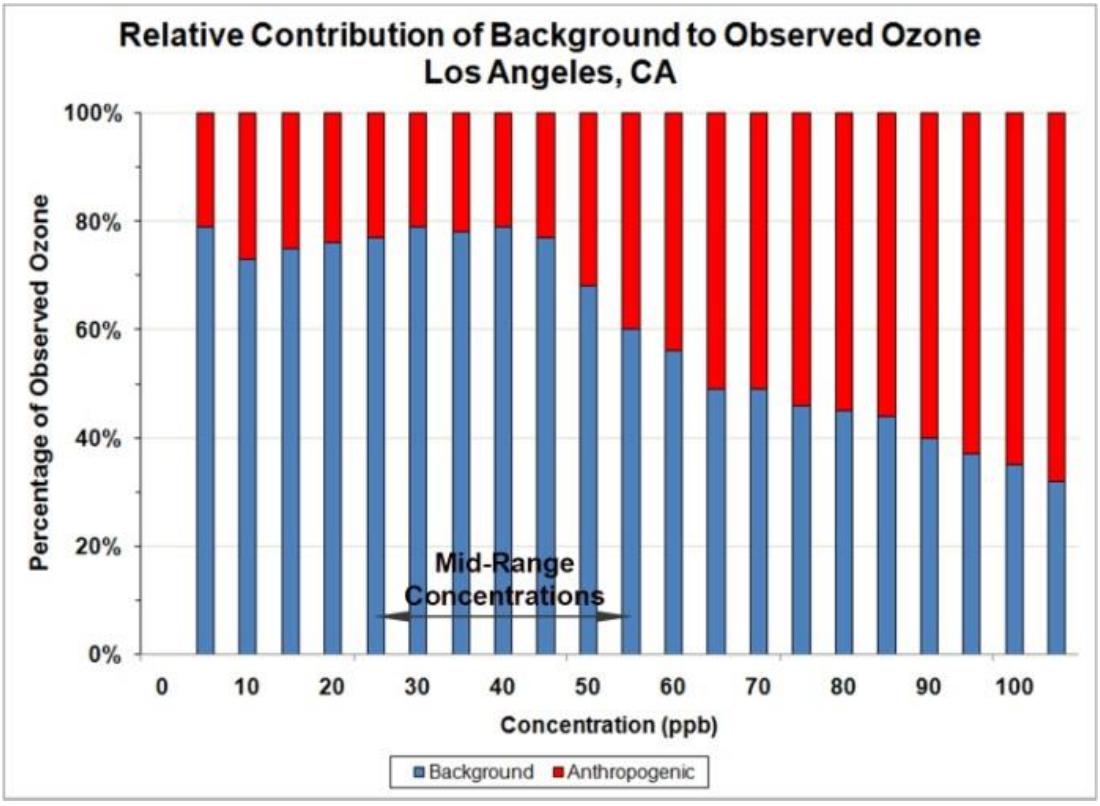


Figure 3-32. Average relative contributions of current hourly background (blue) and anthropogenic O_3 (red) for Los Angeles (CA) area (AQS ID 060719004) in 2006. (Source: Lefohn et al., 2014).

On page 2-23 of the EPA's PA (EPA, 2014a), Fig. 3-33 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 an especially important role in both the low and mid-range total O_3 concentrations (EPA, 2014a).

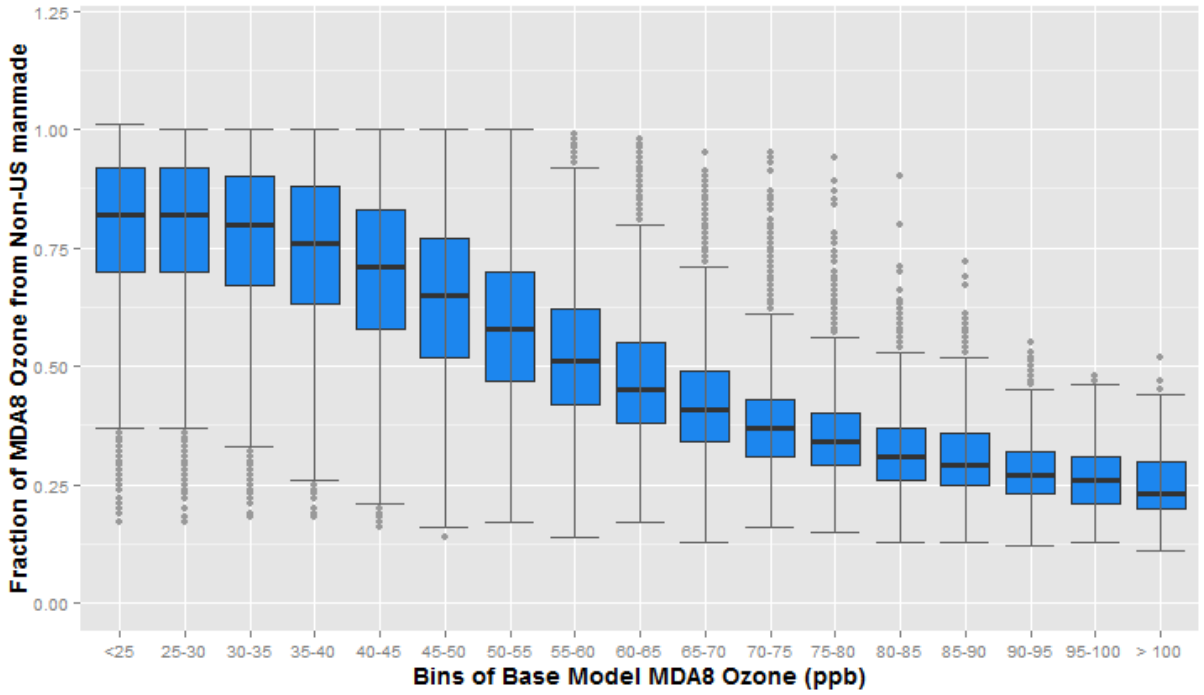
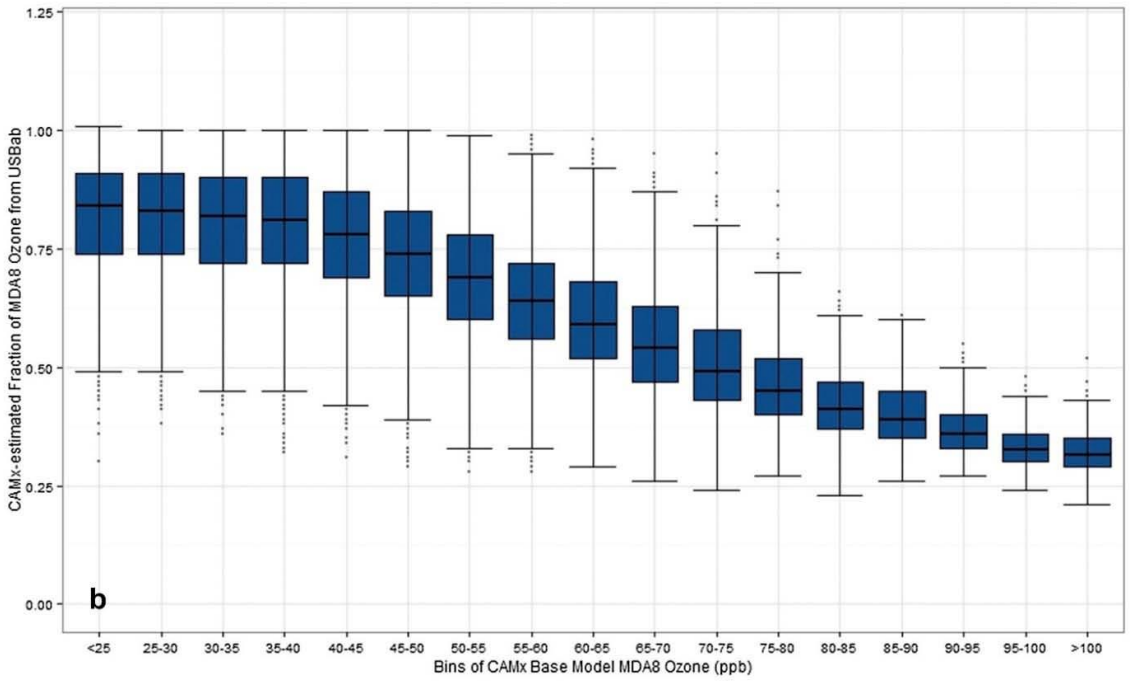


Figure 3-33. Distributions of the relative proportion of apportionment-based U.S. Background (USB_{AB}) to total O_3 (all site-days), binned by modeled MDA8 from the 2007 source-apportionment simulation. Source: Fig. 2-15 in EPA (2014a) with slight modification.

Fig. 3-34 below (Fig. 1-18b on Page 1-68 of 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 O_3 concentration ranges. Similar to the results presented in EPA's 2014 PA (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 3-34. CAMx estimates of daily distributions of bias-adjusted apportioned-based (USB_{AB}) O_3 fraction at monitoring locations across the western U.S. for the period April−October 2007, binned by base model MDA8 O_3 concentration ranges. Source: Figure from Dolwick et al. (2015) as reproduced in the ISA (EPA, 2020a) – page 1-68 Fig. 1-18b.

Lefohn et al. (2014) described a decreasing predicted relative contributions of background O_3 to total O_3 (identified by the authors as "Emissions Influenced Background") 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 (Lefohn et al., 2014; Guo et al., 2018; Dolwick et al., 2015). However, model results do show increasing USB_{AB} and NAB predicted relative contributions of background O_3 to total O_3 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

Depending upon the part of the distribution of hourly average O_3 concentrations, U.S. background O_3 accounts for an important 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 ISA (EPA, 2020a), 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-14 of the ISA).

On Page IS-14 of the ISA, the authors note that tropospheric O_3 derived from stratosphere-troposphere dynamics was described in detail in the 2013 Ozone ISA (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 (EPA, 2013). The 2013 Ozone ISA (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 U.S. 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 in detail 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 the paper by Langford et al. (2009), the authors 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) presented 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 concentrations exceeding 100 ppb were subsequently measured at a surface site in Boulder, and daily maximum 8-h 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 U.S. 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 U.S. 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 U.S., 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 U.S. 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 counts, 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. 3-35 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 counts 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).

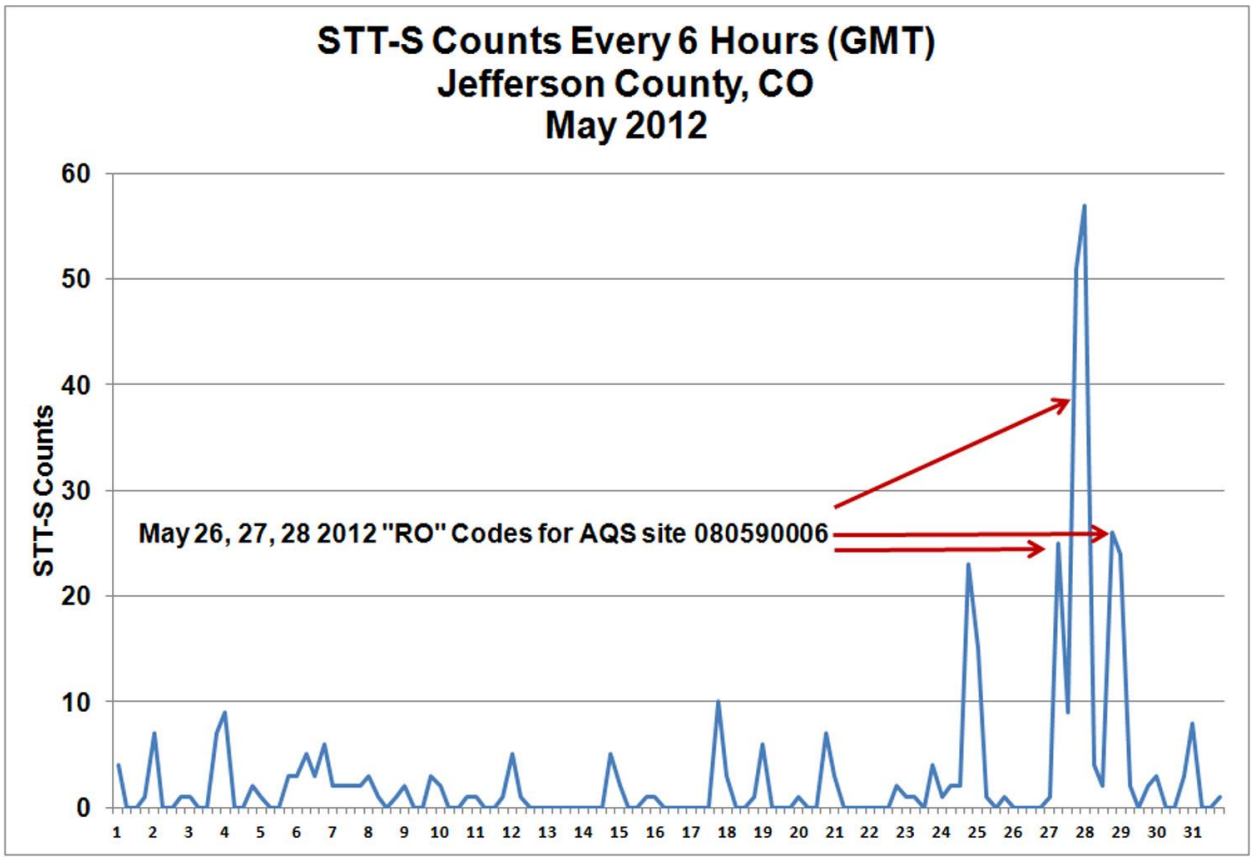


Figure 3-35. Relating STT-S counts for every six hours (GMT) with "RO" codes for a site in Jefferson County, Colorado (AQS ID 080590006) for May 2012. See Lefohn et al. (2011, 2012, 2014) for details how the STT-S values are estimated.

On page IS-15 of the ISA (EPA, 2020a), 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. (EPA, 2013, 2006). 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-15 of the ISA (EPA, 2020a), 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 ISA (EPA, 2020a) 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. However, as discussed later in this subsection, not all high-elevation western U.S. sites have experienced increasing O_3 trends.

One needs to explore further EPA's conclusion that trends at all western high-elevation sites are slowing or reversing due to decreasing East Asian precursor emissions. While emission reductions have occurred in China, O_3 concentrations continue to rise. Lefohn et al. (2017) analyzed trends using hourly average O_3 concentrations from monitoring sites in Hong Kong and mainland China and 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 NO_x 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 NO_x levels may be required before decreasing trends are observed. At many of the Chinese sites, O_3 formation is sensitive to VOCs rather than NO_x; 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 NO_x have increased until recently in mainland China. Conversely, in Hong Kong, there have been large reductions in local emissions of both NO_x 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). NO_x 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.

Xu et al. (2020) presented an integrated analysis of long-term measurements of surface O_3 from eight sites distributed in the North China Plain (NCP) and Yangtze River Delta (YRD), the relatively underdeveloped region Northeast China, and the remote regions in Northwest and Southwest China. Trends and present-day values for seven annual and five seasonal O_3 metrics were characterized through the year 2016. The authors discussed the long-term trends in the various O_3 metrics. Large and significant increases of O_3 were detected at the background site in the NCP, moderate increases at the global baseline site in western China, significant decreases at

the northwestern edge of China, and nearly no trend at other sites. The summer O_3 metrics at the Shangdianzi background site in the NCP indicate increases at rates of more than 2%/yr during 2004–2016. In contrast, O_3 at the Lin'an (LA) background site in the YRD was constant over the period 2006–2016. Xu et al. (2020) note that although tropospheric NO_2 has been declining over the major air pollution regions in China, strong increases of O_3 at many of urban sites in China were observed during 2013–2017 (Lu et al., 2018). The authors noted that this implied that China's policy of emissions reduction in past years was unfavorable for controlling O_3 levels at urban sites, though it has stopped the increases of O_3 at some background sites and effectively lowered $PM_{2.5}$ concentrations.

Long-range transport from Asia has not influenced trend patterns at *all* western U.S. 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 anticipated 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 did not experience increasing trends over the period 2000-2014. Using TOAR data (Schultz et al., 2017), Table 3-2 illustrates the trend patterns for spring (March-May) and summer (June-August) for 13 O_3 monitoring sites. Although not high-elevation sites, Glacier National Park (MT) and Denali National Park (AK) do provide additional information concerning the effects of long-range transport from Asia on O_3 exposures. Oltmans et al. (2010) observed an O_3 episodic enhancement during April 2008 from biomass burning effluent from Eurasia that resulted in unusually high O_3 readings for this time of year in the western U.S. At Denali National Park in central Alaska, an hourly average of 79 ppb was recorded during an 8-h period in which the 8-h average was over 75 ppb, exceeding the O_3 ambient air quality standard threshold value in the U.S. The 8-h daily maximum at Yellowstone on 19 April (i.e., 69 ppb) suggests an enhancement during the period of suspected plume influence of 5-10 ppb above the other relatively high naturally caused O_3 values observed at the WY site. In Table 3-2, the nonparametric Mann-Kendall (M-K) test was used for testing for trends (see Lefohn et al., 2018 for additional information on the use of the M-K statistical method).

Table 3-2. Spring and summer trend patterns for the 4th highest daily maximum 8-h concentration exposure for 13 O_3 monitoring sites in the West for the March-April-May (MAM) and June-July-August (JJA) periods for 2000-2014.

Site	Site ID	Latitude	Longitude	Elev. (m)	MAM Trend	JJA Trend
Gothic	080519991	38.9564	-106.99	2926	No	Negative
Glacier NP	300298001	48.5103	-114	964	No	No
Yellowstone NP	560391011	44.5654	-110.4	2430	No	No
Pinedale	560359991	42.9288	-109.79	2388	No	Negative
Rainier NP	530530012	46.7841	-121.74	1615	No	Negative
Lassen Volcanic NP	060893003	40.54	-121.58	1755	No	Negative
Yosemite NP	060430003	37.7133	-119.71	1599	No	Negative
Great Basin NP	320330101	39.0051	-114.22	2058	No	No
Mesa Verde NP	080830101	37.1984	-108.49	2170	No	No
Denali NP	020680003	63.7232	-148.97	663	No	No

Chiricahua NM	040038001	32.0094	-109.39	1569	No	No
Grand Canyon NP	040058001	36.0586	-112.18	2070	No	Negative
Canyonlands NP	490370101	38.4583	-109.82	1794	No	Negative

NP=National Park
NM=National Monument

The ISA (EPA, 2020a) attributes increasing trend patterns observed until approximately 2010 at high-elevation western U.S. sites to long-range transport from Asia. However, as discussed above, this statement does not appear to be true for all high-elevation western U.S. sites. Additional emission reductions occurring in the future in Asia may have little influence on trends patterns at some western high-elevation O_3 monitoring sites.

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 1-25, the 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; EPA, 2013; Lin et al., 2012a).

Figures 3-36, 3-37, 3-38, 3-39, and 3-40 illustrate as examples the daily maximum 8-h average concentration (MDA8) USB_{AB} estimates, observed daily MDA8 values, and the daily STT-S counts 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 counts 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 (i.e., total observed O_3 and USB_{AB}) 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 total O_3 and USB_{AB} concentrations) occur from mid-May through September, indicating the apparent influence of anthropogenic sources. STT-S events occur across the U.S. 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 U.S. USB_{AB}, while important in the high-elevation sites in the western U.S., is also important at low-elevation sites across the U.S. (Lefohn et al., 2011, 2012, 2014).

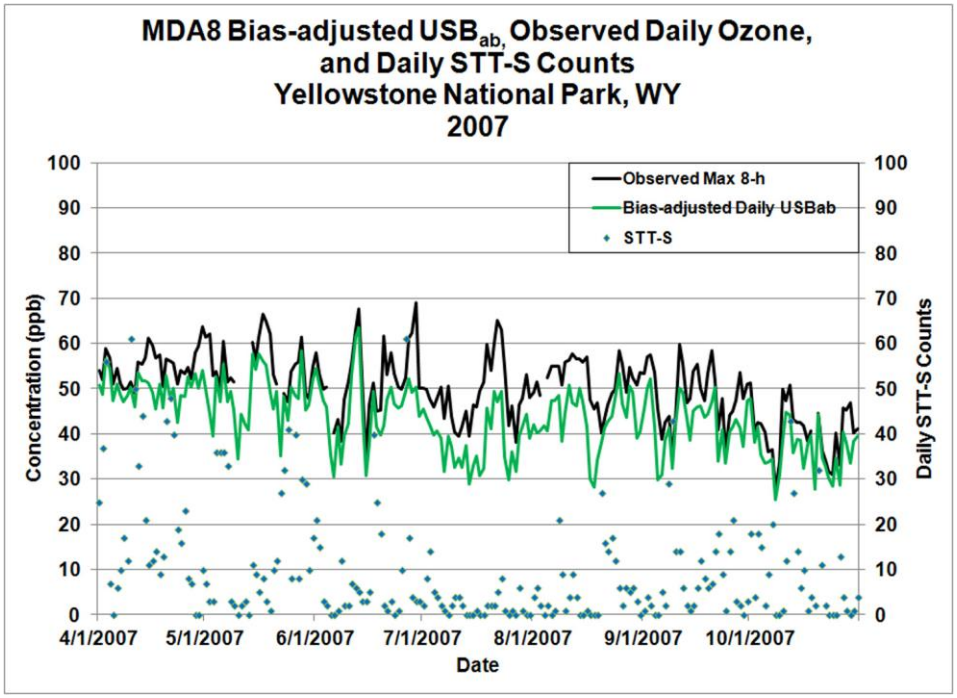


Figure 3-36. A comparison of the observed 8-h daily maximum concentration with the estimated bias-adjusted U.S. Background (USB_{AB}) 8-h daily maximum concentrations for Yellowstone National Park (WY) (AQS ID 560391011) for April-October 2007. The daily stratospheric-tropospheric transport to surface (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.

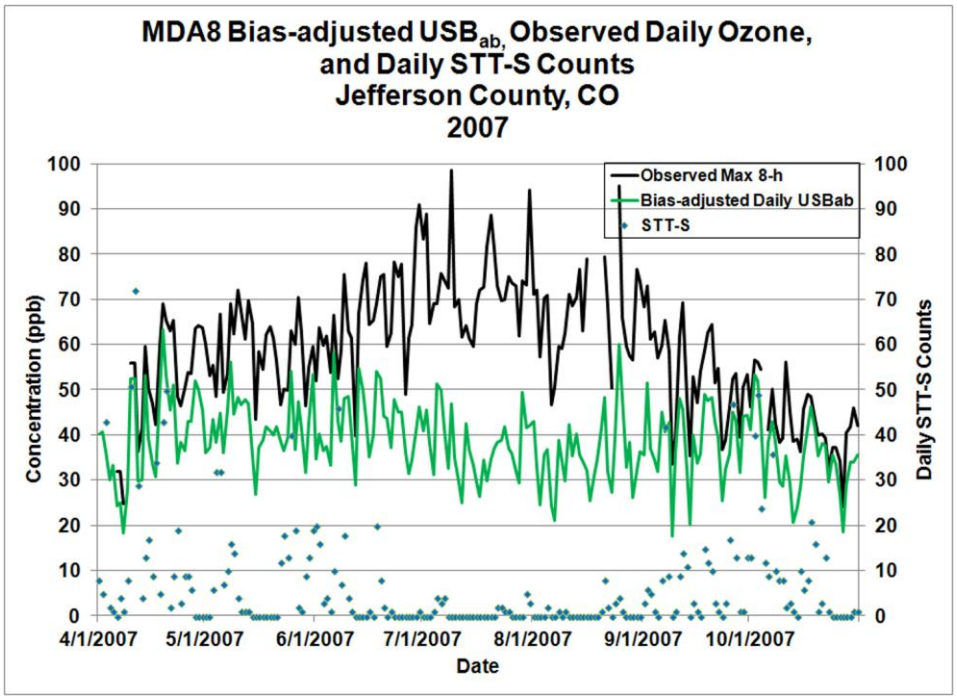


Figure 3-37. A comparison of the observed 8-h daily maximum concentration with the estimated bias-adjusted U.S. Background (USB_{AB}) 8-h daily maximum concentrations for Jefferson County (CO) (AQS ID 080590006) for April-October 2007. The daily stratospheric-tropospheric transport to surface (STT-S) 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.

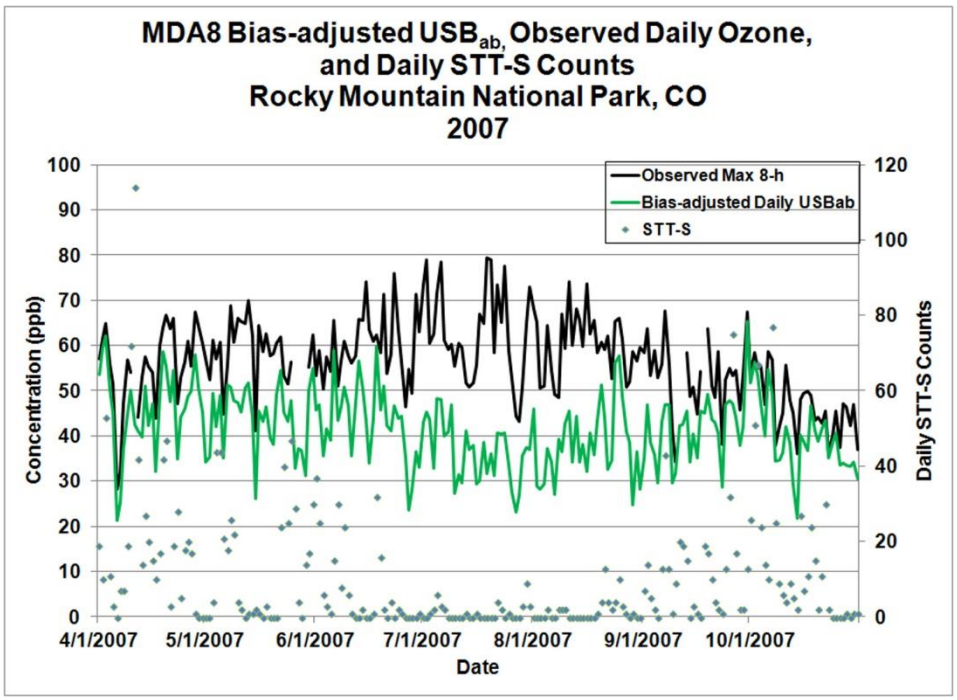


Figure 3-38. A comparison of the observed 8-h daily maximum concentration with the estimated bias-adjusted U.S. Background (USB_{AB}) 8-h daily maximum concentrations for Rocky Mountain National Park (CO) (AQS ID 080690007) for April-October 2007. The daily stratospheric-tropospheric transport to surface (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.

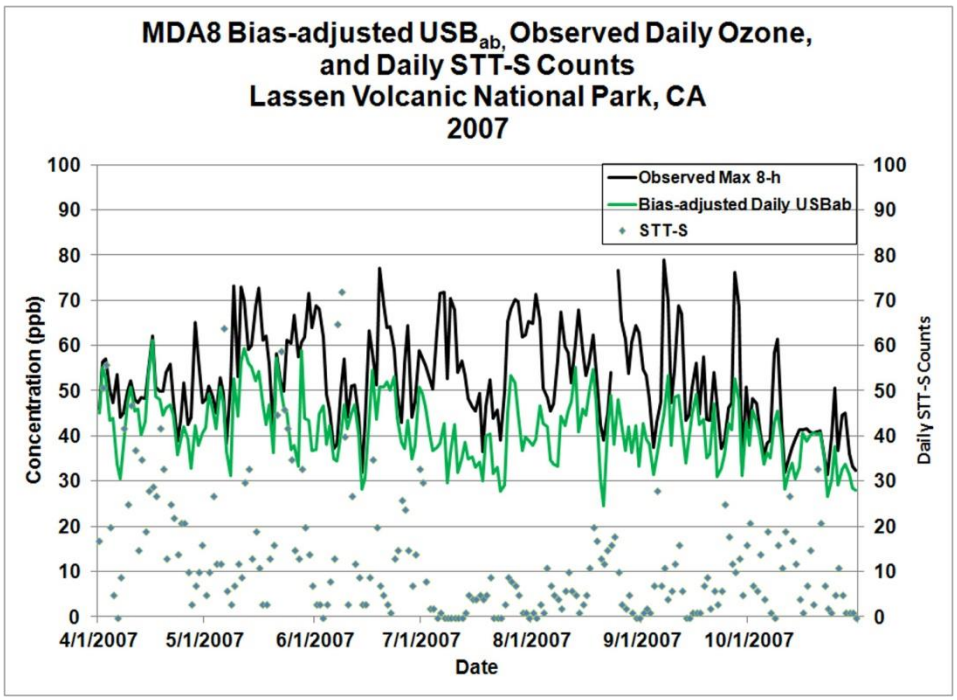


Figure 3-39. A comparison of the observed 8-h daily maximum concentration with the estimated bias-adjusted U.S. Background (USB_{AB}) 8-h daily maximum concentrations for Lassen Volcanic National Park (CA) (AQS ID 060893003) for April-October 2007. The daily stratospheric-tropospheric transport to surface (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.

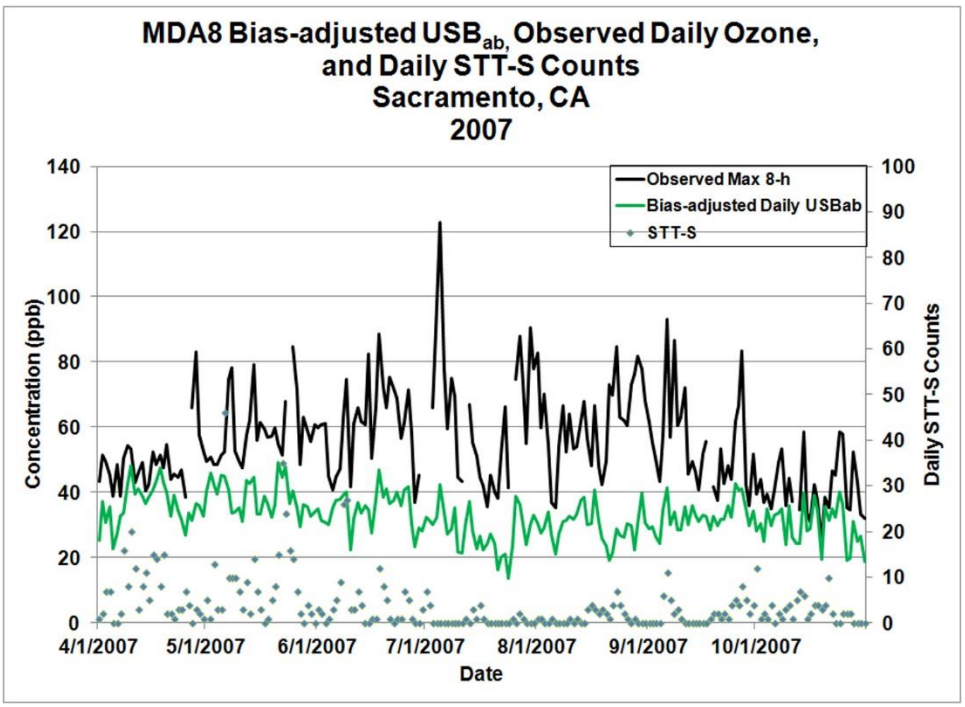


Figure 3-40. A comparison of the observed 8-h daily maximum concentration with the estimated bias-adjusted U.S. Background (USB_{AB}) 8-h daily maximum concentrations for Sacramento (CA) (AQS ID 060670012) for April-October 2007. The daily stratospheric-tropospheric transport to surface (STT-S) trajectories, 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 Observed Ozone Exposure Patterns and Why the Patterns are Important

There appear to be substantial differences between (1) the most current background O_3 modeling results performed by the EPA and presented in the PA (EPA, 2020b) and (2) the EPA results performed in 2014 PA (EPA, 2014a), as well as other results published in the literature. On pages 2-30 and 2-31 of the 2014 PA (EPA, 2014a), the authors state the following:

> For a variety of reasons, it is challenging to present a comprehensive summary of all the components and implications of background O_3. In many forums the term "background" is used generically and the lack of specificity can lead to confusion as to what sources are being considered. Additionally, it is well established that the impacts of background sources can vary greatly over space and time which makes it difficult to present a simple summary of background O_3 levels. Further, background O_3 can be generated by a variety of processes, each of which can lead to differential patterns in space and time, and which often have different regulatory ramifications. Finally, background O_3 is difficult to measure and thus, typically requires air quality modeling which has inherent uncertainties and

potential errors and biases. That said, EPA believes the following concise and three-step summary of the implications of background O_3 on the NAAQS review is appropriate, as based on previous modeling exercises and the more recent EPA analyses summarized herein. First, background O_3 exists and can comprise a considerable fraction of total seasonal mean MDA8 O_3 and W126 across the U.S. Air quality models can estimate the fractional contribution of background sources to total O_3 in an individual area. The largest absolute values of background (NB, NAB, USB, or apportionment-based USB) are modeled to occur at locations in the intermountain western U.S. and are maximized in the ***spring and early summer seasons*** (emphasis added). Second, the modeling indicates that U.S. anthropogenic emission sources are the dominant contributor to the majority of modeled O_3 exceedances of the NAAQS. Higher O_3 days generally have smaller fractional contributions from background. This finding indicates that the relative importance of background O_3 would increase were O_3 concentrations to decrease with a lower level of the O_3 NAAQS. Third and finally, while the majority of modeled O_3 exceedances have local and domestic regional emissions as their primary cause, there can be events where O_3 levels approach or exceed 60-75 ppb due to the influence of background sources. These events are relatively infrequent and EPA has policies that could allow for the exclusion of air quality monitoring data affected by these types of events from design value calculations.

To summarize the EPA's key observations in the 2014 PA (EPA, 2014a) about its modeling results stated

- First, background O_3 exists and can comprise a considerable fraction of total seasonal mean MDA8 O_3 and W126 across the U.S. Air quality models can estimate the fractional contribution of background sources to total O_3 in an individual area. The largest absolute values of background (NB, NAB, USB, or apportionment-based USB) are modeled to occur at locations in the intermountain western U.S. and are maximized in the spring and early summer seasons.

- Second, the modeling indicates that U.S. anthropogenic emission sources are the dominant contributor to the majority of modeled O_3 exceedances of the NAAQS. Higher O_3 days generally have smaller fractional contributions from background. This finding indicates that the relative importance of background O_3 would increase were O_3 concentrations to decrease with a lower level of the O_3 NAAQS.

- Third and finally, while the majority of modeled O_3 exceedances have local and domestic regional emissions as their primary cause, there can be events where O_3 levels approach or exceed 60-75 ppb due to the influence of background sources. These events are relatively infrequent and EPA has policies that could allow for the exclusion of air quality monitoring data affected by these types of events from design value calculations.

The seasonal pattern for background O_3 mentioned in the first bullet is supported by results summarized in the 2013 ISA (EPA, 2013). On page 2-17 of the 2014 PA (EPA, 2014a), the authors note

> The ISA (EPA 2013, section 3.4) previously established that background concentrations vary spatially and temporally and that simulated mean background concentrations are highest at high-elevation sites within the western U.S. ***Background levels typically are greatest over the U.S. in the spring and early summer*** (emphasis added).

As noted in Section 3.2.4 of this review, the modeling results described in the PA (EPA, 2020b) indicate that

- The current analysis indicates that natural and USA O_3 contributions peak during the traditional O_3 season (May through September), while long-range intercontinental transport of international O_3 (i.e. contributions from China, India etc.) peaks in the spring (February through May). (page 2-64).

- The Natural component of USB exhibits the largest magnitude difference between the West and East. (page 2-65).

- The natural contribution has a single maximum in late summer in the West, whereas, in the East there is evidence of two peaks— the largest in late Spring and a second peak in early Fall. (page 2-48).

The EPA's USB modeling results summarized in the PA (EPA, 2020b) appear to show a different seasonal pattern for when background O_3 is highest when these results are compared with the previous conclusions in the 2014 PA (EPA, 2014a). As noted in the PA (EPA, 2020b), the USA contributions (i.e., anthropogenic) that drive exceedances generally peak in summer. As noted in the first bullet above, the Natural component in the model described in the PA (EPA, 2020b) also peaks during the traditional O_3 season. In contrast, as pointed out in the 2014 PA (EPA, 2014a), the ISA (EPA 2013, section 3.4) previously established that background concentrations are greatest over the U.S. in the spring and early summer (i.e., March – mid-June). As indicated in Section 3.2.4 of this review, the authors of the ISA (EPA, 2020a) did not resolve these conflicting conclusions identifying, based on background O_3 modeling, when seasonal mean background O_3 is greatest. Unfortunately, the PA (EPA, 2020b) did not adequately discuss these inconsistencies. However, a great majority of the peer-reviewed literature has identified spring and early summer as the period when background O_3 concentrations are the greatest across the U.S. In a recent review of the literature, Jaffe et al. (2018) concluded that model-calculated USB O_3 was greatest during the March through June period.

In the EPA's AQS database there is information that can provide guidance for better understanding the distribution of hourly average background O_3 concentrations, as well as the seasons that exhibit the highest background O_3 concentrations. In the database, information from some of the O_3 monitoring sites illustrate the compression described in the literature about the

distribution of hourly average O_3 concentrations, where the highest average values shift downward toward the mid-range concentrations and the lowest average concentrations shift upwards toward the mid values. In many cases for inland monitoring sites, the resulting distribution of hourly average concentrations resembles a bell-shaped-like curve.

The EPA, in cooperation with the U.S. Forest Service, established a network of air monitoring stations (referred to as National Air Pollution Background Network (NAPBN)), which was designed to measure levels of O_3 in remote areas within the contiguous 48 states (Evans et al., 1983). There were 8 monitoring sites (Green Mt. NF, VT; Kisatchie NF, LA; Custer NF, MT; Chequamegon NF, WI; Mark Twain NF, MO; Croatan NF, NC; Apache NF, AZ; Ochoco NF, OR) at various National Forests, which measured O_3, wind speed and direction, temperature, relative humidity, and solar radiation. The network was established to provide a reasonable long-term and continuous record of O_3 concentrations and patterns in areas well removed from anthropogenic sources of air pollution and to make these data available to the EPA and other interested researchers.

Of the 6 NAPBN stations operational for the entire year of 1979, all but the site at Custer NF in Montana recorded hourly average O_3 concentrations which were more than 0.08 ppm. Most days with elevated O_3 concentration, as defined by hourly average values >0.08 ppm, occurred in the spring and early summer months. Evans et al. (1983) hypothesized that the spring events, which occurred at the 7 NAPBN stations, may have been associated with natural sources (i.e., the stratosphere). The Custer NF site was located at Fort Howes in the eastern plains of Montana, near the Wyoming border. Fort Howes is located about 32 km south of Ashland, Montana. Inspecting the hourly average concentration data for the Custer NF site (1250 m, 45° 14' 00" N, 106° 15' 00" W), Fig. 3-41 illustrates that the frequency distribution appears to approach. a Gaussian-like (i.e., bell shaped) distribution. Five hourly average concentrations of 75 ppb occurred on 25 April 1979. There were 16 hourly average concentrations at 70 ppb at the site, which occurred on 17 April (4 occurrences), 19 April (2 occurrences), 25 April (5 occurrences), and 26 April (5 occurrences). The two hourly instances on 19 April occurred at 0000 and 0100 in the early morning hours local standard time (LST). The 5 hourly instances of 70 ppb on 26 April occurred during the early morning hours of 0500-0900 LST. In other words, at the Montana site all the maximum hourly average O_3 concentrations occurred during the springtime and appear to have been related to possible stratospheric-to-tropospheric transport to the surface (STT-S). Without more detailed information concerning the meteorological conditions during the April 1979 periods, it is not possible to definitively associate the highest hourly exposures with natural stratospheric intrusions. However, it is a reasonable explanation for the cause of the elevated hourly O_3 concentrations and tends to support the hypothesis stated in Evans et al. (1983).

Lefohn et al. (1998) compared the Custer NF bell-shaped-like frequency distribution (Fig. 3-41) with the distribution of hourly average concentrations for an urban influenced site in Jefferson County, KY (AQS ID 211110027). The frequency distribution of the hourly average O_3 concentrations at the Kentucky site appeared to have a more log-normal-like shape (Fig. 3-42). In contrast to the Custer NF site, the urban-influenced site in Kentucky showed frequent high and low hourly average concentrations. Lefohn et al. (1998) noted that the Kentucky site

appeared to be influenced by NO titration of O_3 because of the occurrence of more frequent low hourly average concentrations.

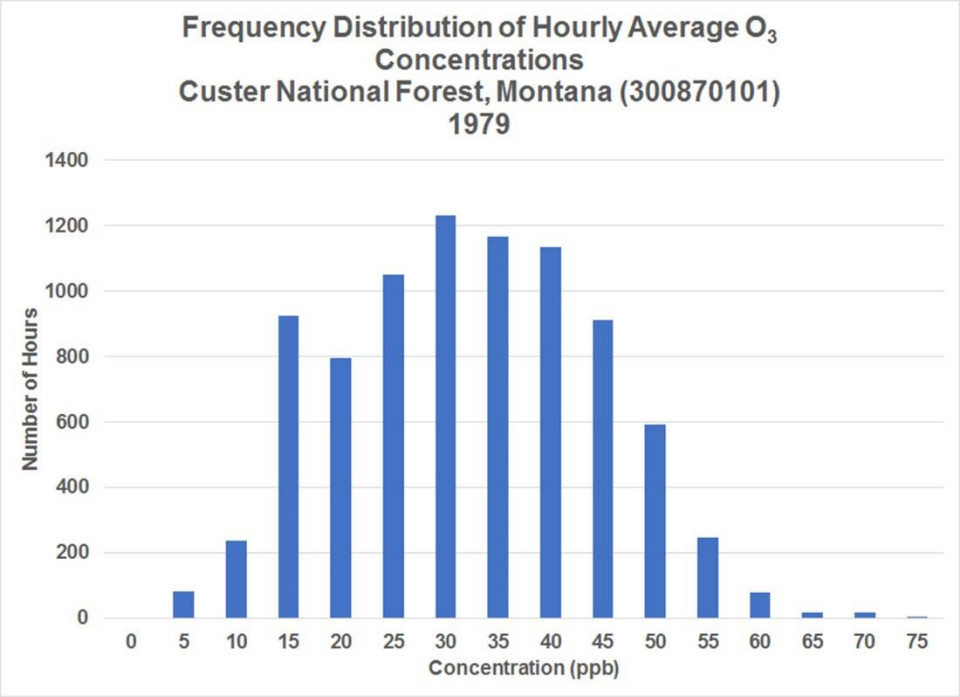


Figure 3-41. Frequency distribution of the hourly average O_3 concentrations in 1979 for Custer National Forest (MT) (300870101) monitoring station. Source of data is from the EPA's AQS database.

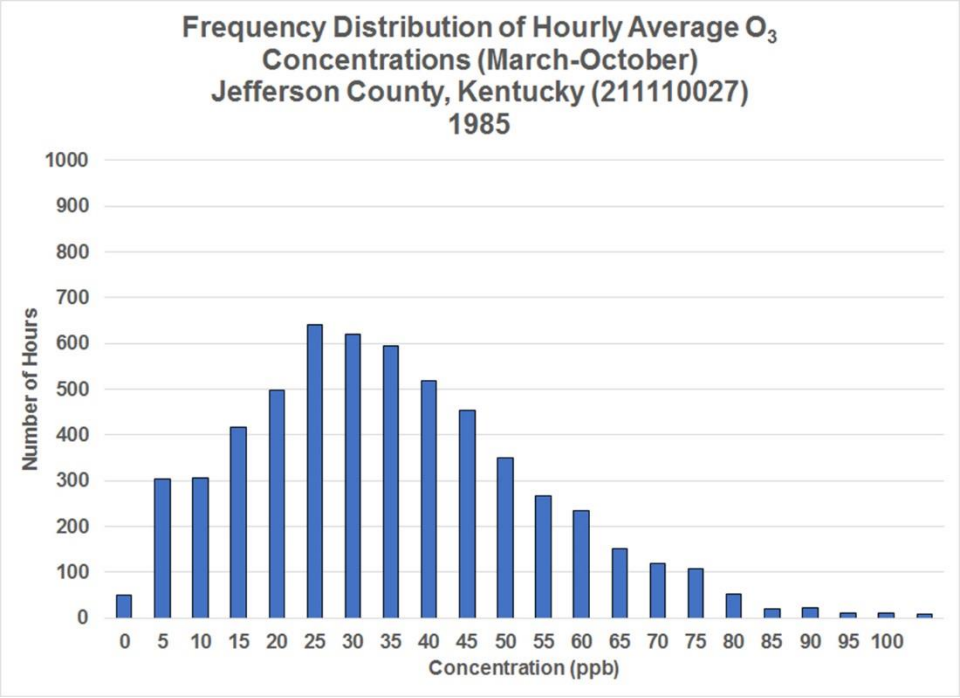


Figure 3-42. Frequency distribution of the hourly average O_3 concentrations in 1985 for an urban-influenced site in Jefferson County (KY) (211110027) monitoring station. Source of data is from the EPA's AQS database.

Based on the comparison of the two sites, Lefohn et al. (1998) hypothesized that as adequate control strategies were implemented to meet the O_3 NAAQS that the distribution pattern of hourly average concentrations for inland monitoring sites might approach the distribution pattern observed at the Montana site, as well as other remote sites in the western U.S. The distribution of hourly average data for the same Kentucky site for 2017 is shown in Fig. 3-43. The distribution shape in 2017 is similar to the bell-shaped-like distribution observed for the Custer NF site in 1979. Although the site is still influenced by anthropogenic sources, the highest hourly average O_3 concentration has been reduced from 112 ppb (experienced in 1985) to 77 ppb (experienced in 2017). In addition, reviewing the two distribution figures for Jefferson County, Kentucky, illustrates the compression, where the highest hourly average concentrations moved downward toward the mid-range values and the lowest concentrations shifted upward toward the mid-range values.

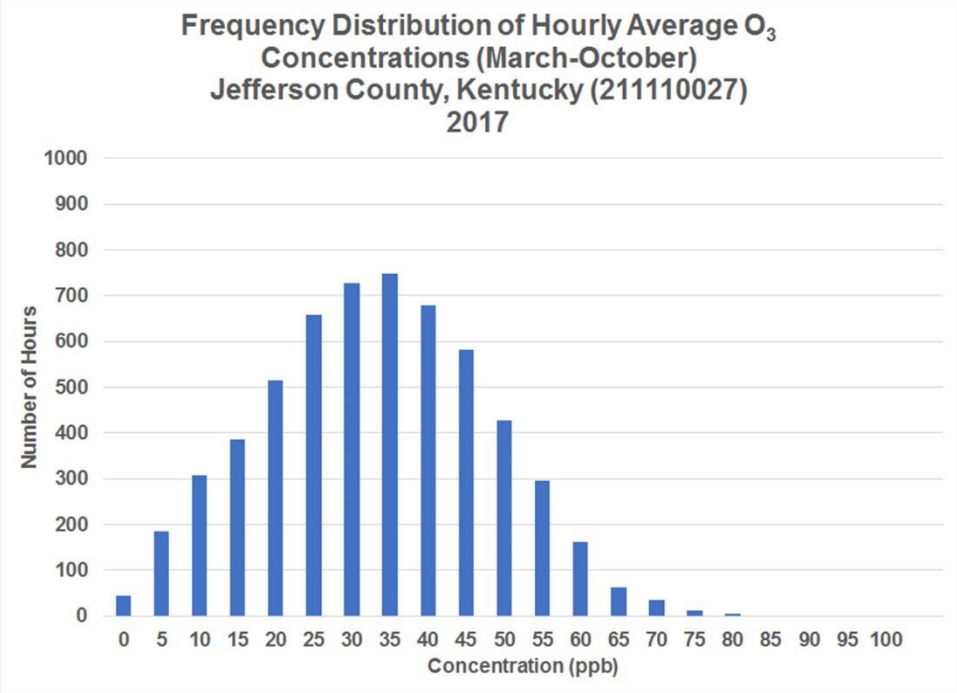


Figure 3-43. Frequency distribution of the hourly average O_3 concentrations in 2017 for an urban-influenced site in Jefferson County (KY) (211110027) monitoring station. Source of data is from the EPA's AQS database.

Building upon the observations noted by Lefohn et al. (1998), EPA (2014a), and Simon et al. (2015), Lefohn et al. (2017) (authors: A. Lefohn, C. Malley, H. Simon, B. Wells, X. Xu, L. Zhang, and T. Wang) used data from 481 sites (276 in the EU, 196 in the U.S., and 9 in China) to investigate the response of 14 human health and vegetation O_3 exposure metrics to changes in hourly O_3 concentration distributions over time that resulted from changes in emissions. For the U.S. sites used in the analysis, the following selection criteria were used: (1) sites had to collect

data with a minimum of 20 years (1995-2014); sites initiating monitoring as far back as 1980 were also used; (2) sites had to collect data year-round over the entire period; and (3) sites had to not experience large data gaps in the monitoring record with more than one large data gap of up to 1 year in length. The authors reported that at a majority of EU and U.S. sites, there was a reduction in the frequency of both relatively high and low hourly average O_3 concentrations. The patterns of changes in hourly average O_3 concentration distributions were separated into ten distinct 'trend type' categories. For each site, Lefohn et al. (2017) identified what portion of the distribution of hourly average concentrations had shifted. For characterizing patterns of change for the distributions, the trend types were described as follows (yellow highlights are provided to assist the reader):

- **Trend Type 0:** No trend.

- **Trend Type 1**: Both ends of the distribution shift toward the center. (Decreasing frequency of high and low concentrations).

- **Trend Type 2**: Low end shifts upward but high end does not change. (Decreasing frequency of low concentrations; increasing frequency of middle concentrations).

- **Trend Type 3**: High end shifts downwards but no change at lower end (Decreasing frequency of high concentrations; increasing frequency of middle concentrations).

- **Trend Type 4**: Entire distribution shifts downwards (Decreasing frequency of high concentrations, increasing frequency of low concentrations).

- **Trend Type 5**: The distribution shifts from the center toward both the high and the low ends of the distribution. (Increasing frequency of high and low concentrations).

- **Trend Type 6**: The middle of the distribution shifts downward but the high end does not change. (Increasing frequency of low concentrations, decreasing frequency of middle concentrations).

- **Trend Type 7**: The middle of the distribution shifts upward but the low end does not change. (Increasing frequency of high concentrations, decreasing frequency of middle concentrations).

- **Trend Type 8**: Entire distribution shifts upwards. (Increasing frequency of high concentrations, decreasing frequency of low concentrations).

- **Trend Type X**: Complex trends that do not fall into any of the categories listed above. It is not possible to categorize portions of the O_3 distribution into "low", "middle", and "high" for this trend type because the directions of the trends shift more than two times across the distribution.

Trend Type 1 (highlighted in yellow above), as discussed in Lefohn et al. (2017), identified the number of sites that exhibited the compression pattern described in Lefohn et al.

(1998), EPA (2014a), and Simon et al. (2015). Lefohn et al. (2017) noted that because relative shifts of low and high hourly concentrations within the Trend Type 1 distribution can influence the median concentration, Trend Type 1 sites were further grouped in their analyses into three subcategories based upon trends in the median concentration: (1) "1a" sites had increasing trends in the median; (2) "1b" sites had no trend in the median; and (3) "1c" sites had decreasing trends in the median.

Table 3-3 (reproduced from Table 2 in Lefohn et al., 2017) below summarizes the trend type assignments for the 196 U.S. monitoring sites. Trend Type 1 (i.e., compression of the high- and low-end concentrations within the distribution, shifting more O_3 concentrations toward the center) was the most predominant trend pattern (84% of U.S. sites (165)). Most of the U.S. Trend Type 1 sites were classified as Trend Type 1a (i.e., increasing median); almost 30% were classified as Trend Type 1b (i.e., no trend in the median). Sixty-one percent of the U.S. sites analyzed by Lefohn et al. (2017) exhibited (1) compression of the high and low concentrations toward the middle of the distribution of hourly average O_3 concentrations and (2) increasing trends in the median concentration. It would be anticipated that the observed increase in median concentrations would result in the average concentration also increasing for many of the same sites.

Table 3-3. Number of U.S. sites in each trend type category from the scatter dataset by degree of urbanization. Values in parentheses indicate the percent of rural, suburban, or urban sites that fall into each category. Source: Lefohn et al. (2017).

Trend Type	Rural	Suburban	Urban	Total
0 (No trend)	5 (6%)	0 (0%)	0 (0%)	5 (3%)
1a	31 (36%)	44 (76%)	44 (86%)	119 (61%)
1b	26 (30%)	11 (19%)	5 (10%)	42 (21%)
1c	4 (5%)	0 (0%)	0 (0%)	4 (2%)
2	4 (5%)	1 (2%)	2 (4%)	7 (4%)
3	7 (8%)	1 (2%)	0 (0%)	8 (4%)
4	9 (10%)	1 (2%)	0 (0%)	10 (5%)
7	1 (1%)	0 (0%)	0 (0%)	1 (<1%)
Total	87	58	51	196

The observations from Lefohn et al. (1998), EPA (2014a), Simon et al. (2015), Lefohn et al. (2017), and Lefohn et al. (2018) indicate that emission reductions have resulted in some sites experiencing a compression of the distribution of hourly average O_3 concentrations and, in some cases, the compression of the high and low concentrations toward the middle results in a bell-shaped-liked distribution. ***As emission reductions reduce the absolute value of the higher concentrations, background O_3 increases its role in the percent contribution to the total O_3 measured. Mathematically stated, in the limit, as U.S. anthropogenic contributions approach zero (which is not possible socially), what remains is a distribution of hourly average O_3***

concentrations that represent background O_3 that, in many cases for inland sites, is described as a bell-shaped-like curve.

Several examples exist today that illustrate the bell-shaped-like distribution described above. The amplitude and the width of the distribution vary from site to site. Fig. 3-44 illustrates the distribution of hourly average O_3 concentrations for 2018 for the Yellowstone National Park (WY) site. As indicated previously, the Yellowstone NP site exhibited no trend (using the nonparametric Mann-Kendall statistical test). For the year 2006, Lefohn et al. (2014) reported that background O_3 contributed a major portion of the total observed hourly average O_3 concentrations for all concentrations. Fig. 3-45 illustrates the distribution of hourly average O_3 concentrations for a site at Mesa Verde (CO) National Park. This site also exhibits the bell-shaped-like distribution pattern. The change in distribution patterns for the hourly average O_3 concentrations are not just occurring in the western U.S. Figures 3-46 and 3-47 illustrate the distribution pattern for a site located in Garrett County, Maryland for 2005 and 2018, respectively. The site is identified in the AQS database as rural forested. In 2005, the Garrett County site experienced a maximum hourly average O_3 concentration of 100 ppb. In 2018, the site experienced a maximum hourly value of 75 ppb. Fig. 3-48 illustrates the distribution pattern for a site in Monroe County, Missouri. The site setting is listed as rural in the EPA's AQS database. In 2000, the Monroe County site experienced a maximum hourly average O_3 concentration of 91 ppb. In 2018, the site exhibited a maximum hourly average O_3 concentration of 76 ppb.

The bell-shaped-like distribution curve provides us with the ability to gain insights about the distribution of background O_3 concentrations site by site. *While we cannot quantify the exact distribution of the hourly average background O_3 concentrations, we know that as the compression occurs as emission are reduced that background O_3 is encapsulated in the distribution.* For example, for the Yellowstone NP (WY) site in 2018 (Fig. 3-44), the highest frequency of hourly average O_3 concentrations is in the range of 40-45 ppb. The highest hourly average O_3 concentrations at this site are most likely associated with stratospheric contributions. The Mesa Verde (CO) site in 2018 (Fig. 3-45) shows the highest frequency of hourly average O_3 concentrations in the range of 45-50 ppb. For the Garrett County (MD) site in 2018 (Fig. 3-47), the highest frequency of hourly average O_3 concentrations is 35 ppb. For the Monroe County (MO) site in 2018 (Fig. 3-48), the maximum frequency of O_3 concentrations is 30 ppb. In summary, for those sites still influenced by anthropogenic sources within the U.S./Canada/Mexico domain, as emissions continue to be reduced, the absolute values of the highest frequency in the distribution will continue to increase as background O_3 becomes even more dominant.

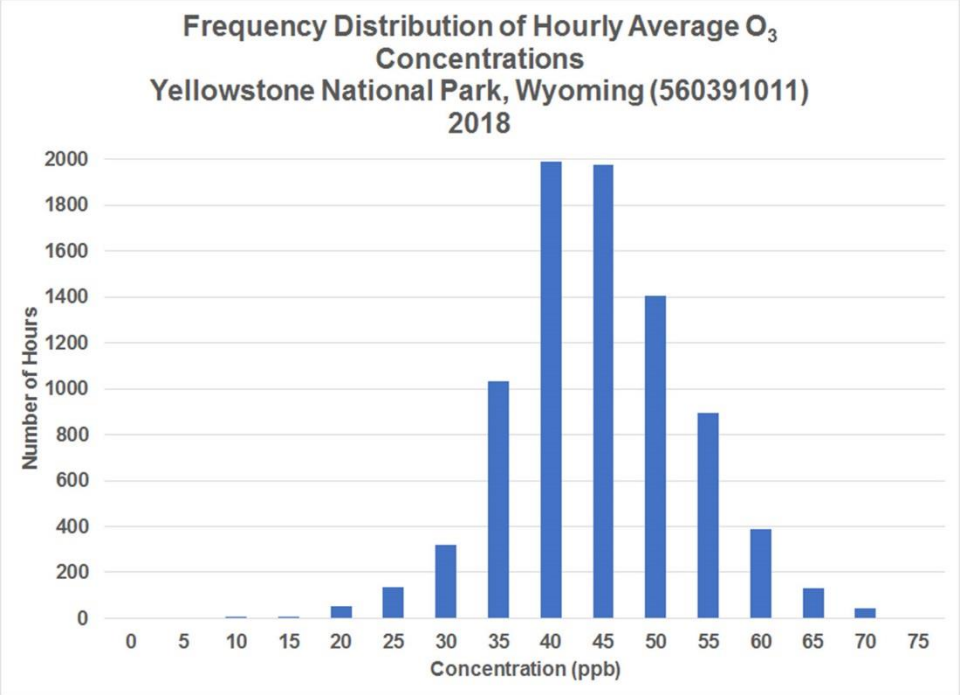


Figure 3-44. Frequency distribution of the hourly average O_3 concentrations for January-December 2018 for Yellowstone NP (WY) (560391011). Source of data is from the EPA's AQS database.

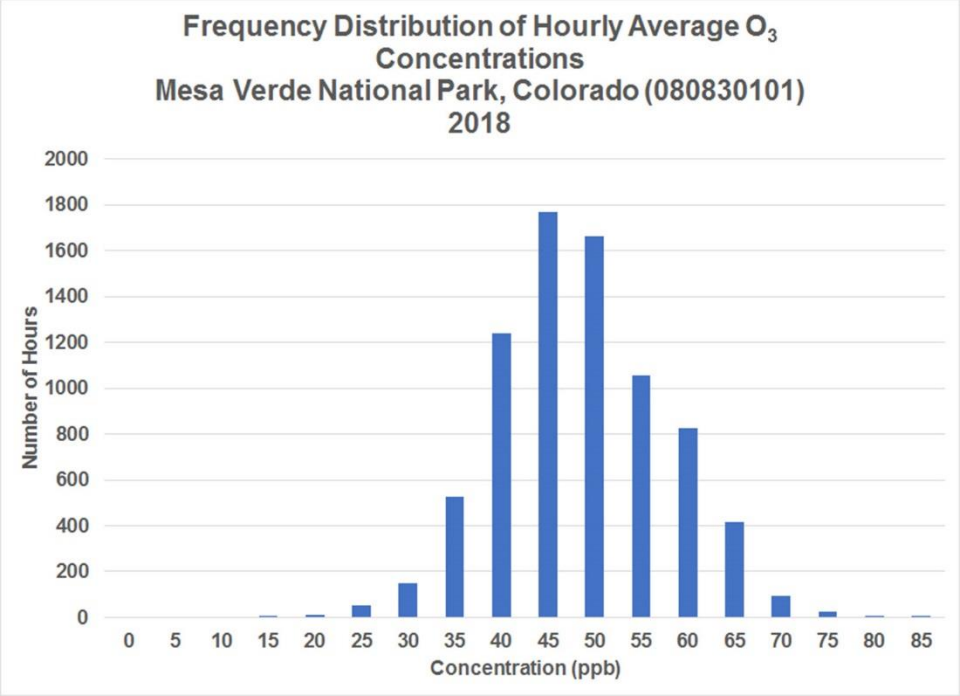


Figure 3-45. Frequency distribution of the hourly average O_3 concentrations for January-December 2018 for Mesa Verde National Park (CO) (080830101). Source of data is from the EPA's AQS database.

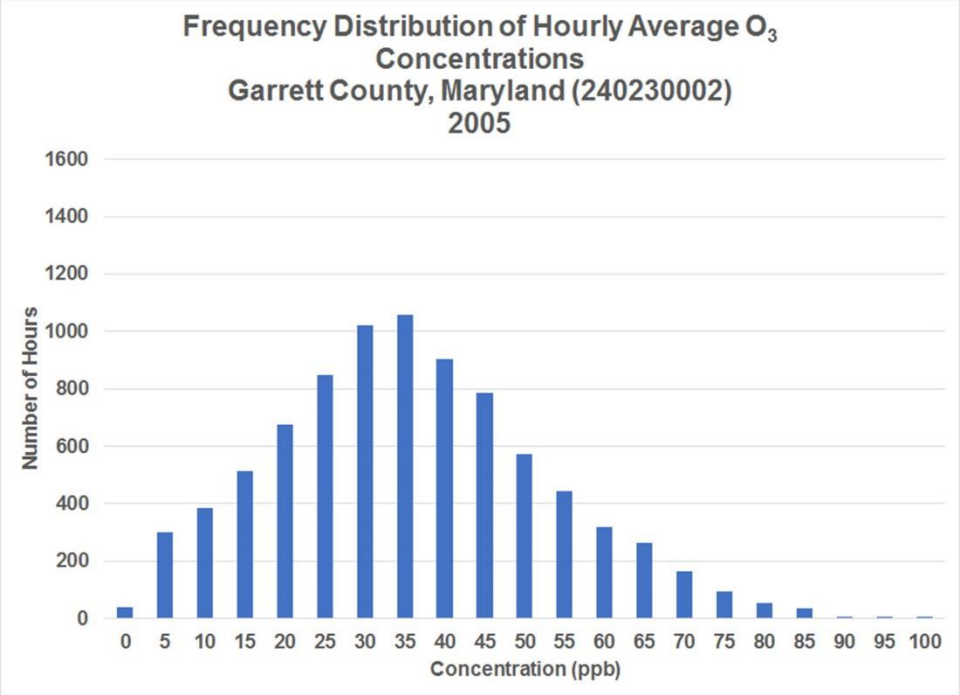


Figure 3-46. Frequency distribution of the hourly average O_3 concentrations for January-December 2005 for Garrett County (MD) (240230002). Source of data is from the EPA's AQS database.

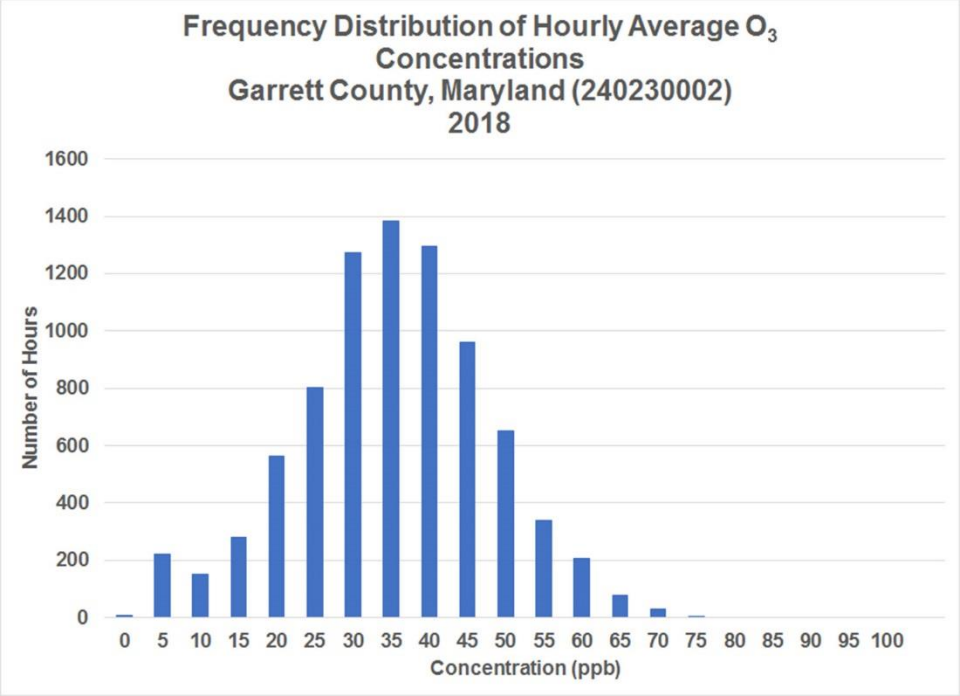


Figure 3-47. Frequency distribution of the hourly average O_3 concentrations for January-December 2018 for Garrett County (MD) (240230002). Source of data is from the EPA's AQS database.

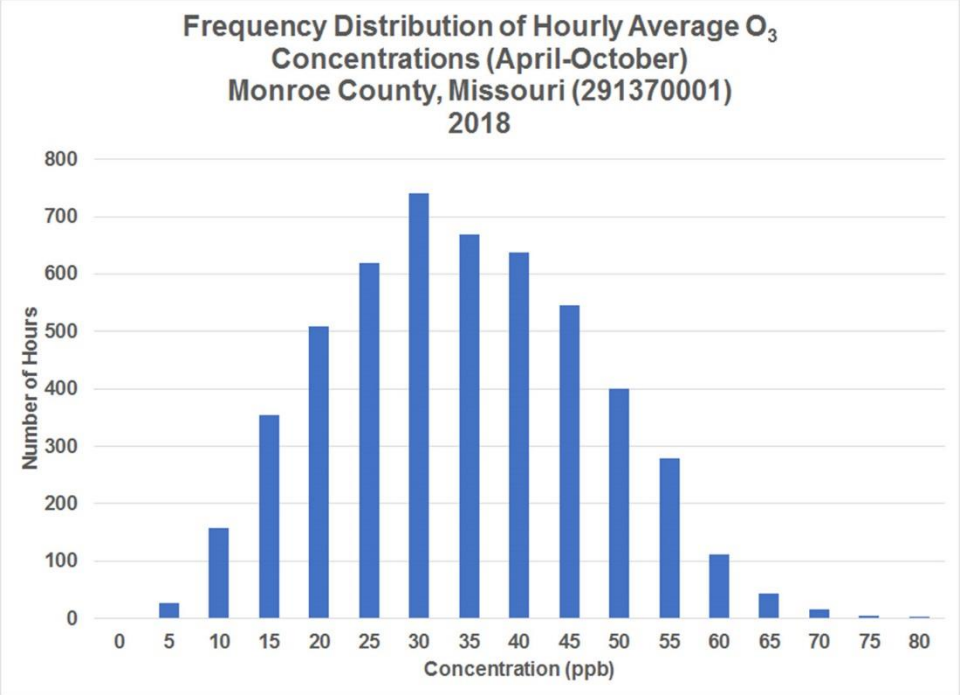


Figure 3-48. Frequency distribution of the hourly average O_3 concentrations for January-December 2018 for Monroe County (MO) (291370001). Source of data is from the EPA's AQS database.

In addition to the observation of a compression of the distribution of hourly average O_3 concentrations (with the higher values shifting downward toward the mid-values and the lower values shifting upward toward the mid concentrations) as emission reductions occur, the period when the higher O_3 exposures occur has shifted from the July-August period toward the March-June months. The in its Health Risk and Exposure Assessment analysis (EPA, 2014b) noted this shift in its risk assessment modeling results. In addition, EPA had concern that the O_3 monitoring season defined for each state and the District of Columbia was not adequately capturing the occurrence of daily maximum 8-h O_3 average concentrations equal to or above 0.060 ppm. In response to this concern, in the 2015 O_3 NAAQS rulemaking (Federal Register, 2015 – page 65416), the EPA determined that the lengthening of the O_3 monitoring seasons in 32 states and the District of Columbia was required. The Agency indicated that ambient O_3 concentrations in these areas could approach or exceed the level of the NAAQS, more frequently and during more months of the year compared with the length of the O_3 seasons prior to 2015. The EPA concluded that it was important to monitor for O_3 during the periods when ambient concentrations could approach the level of the NAAQS to ensure that the public was informed when exposure to O_3 could reach or has reached a level of concern. The EPA completed an analysis to address whether extensions of currently required monitoring seasons were appropriate (Rice, 2014). In the EPA analysis, all available data in the AQS were used, including data from monitors that collected O_3 data year-round during 2010-2013. More than half of the O_3 monitors were voluntarily operated on a year-round basis by monitoring agencies. The Agency determined the number of days where one or more monitors experienced a daily maximum 8-h O_3 average equal to or above 0.060 ppm in the months outside each state's current O_3 monitoring season and the pattern of those days in the out-of-season months. The EPA believed that a threshold of 0.060

ppm, taking into consideration reasonable uncertainty, served as an appropriate indicator of ambient conditions that may be conducive to the formation of O_3 concentrations that approach or exceed the NAAQS. The Agency also considered regional consistency, particularly for those states with little available data. EPA noted that seasonal O_3 patterns varied year-to-year due primarily to highly variable meteorological conditions conducive to the formation of elevated O_3 concentrations early or late in the season in some years and not others. The EPA believed it was important that O_3 monitors operated during all periods when there was a reasonable possibility of ambient levels approaching the level of the NAAQS.

As a result of its analysis, modifications to the O_3 monitoring season involved adding earlier, as well as later months to the monitoring seasons that were used prior to 2015. Changes to the required O_3 monitoring seasons were finalized by the EPA (Federal Register, 2015 – page 65419) as follows for these states and the District of Columbia:

> Colorado: Proposed addition of January, February, October, November, and December is finalized. The required season is revised to January – December.
>
> Connecticut: Proposed addition of March is finalized, revising season to March – September.
>
> Delaware: Proposed addition of March is finalized, revising season to March – October.
>
> District of Columbia: Proposed addition of March is finalized, revising season to March – October.
>
> Florida: Proposed addition of January, February, November, and December is finalized. The required season is revised to January – December.
>
> Idaho: Proposed addition of April is finalized, revising season to April – September.
>
> Illinois: Proposed addition of March is finalized, revising season to March – October.
>
> Indiana: Proposed addition of March and October, revising season to March – October.
>
> Iowa: Proposed addition of March is finalized, revising season to March – October.
>
> Kansas: Proposed addition of March is finalized, revising season to March – October.
>
> Maryland: Proposed addition of March is finalized, revising season to March – October.

Massachusetts: Proposed addition of March is finalized, revising season to March – September.

Michigan: Proposed addition of March and October is finalized, revising season to March – October.

Minnesota: Proposed addition of March is finalized, revising season to March – October.

Missouri: Proposed addition of March is finalized, revising season to March – October.

Montana: Proposed addition of April and May is finalized, revising season to April – September.

Nebraska: Proposed addition of March is finalized, revising season to March – October.

New Hampshire: Proposed addition of March is finalized, revising season to March – September.

New Jersey: Proposed addition of March is finalized, revising season to March – October.

New York: Proposed addition of March is finalized, revising season to March – October.

North Carolina: Proposed addition of March is finalized, revising season to March – October.

North Dakota: Proposed addition of March and April is finalized, revising season to March – September.

Ohio: Proposed addition of March is finalized, revising season to March – October.

Pennsylvania: Proposed addition of March is finalized, revising season to March – October.

Rhode Island: Proposed addition of March is finalized, revising season to March – September.

South Carolina: Proposed addition of March is finalized, revising season to March – October.

South Dakota: Proposed addition of March, April, May, and October is finalized, revising season to March – October.

Texas (Northern AQCR 022, 210, 211, 212, 215, 217, 218): Proposed addition of November is finalized, revising season to March – November.

Utah: Proposed addition of January, February, March, April, October, November, and December is finalized. The required season is revised to January – December.

Virginia: Proposed addition of March is finalized, revising season to March – October.

West Virginia: Proposed addition of March is finalized, revising season to March – October.

Wisconsin: Proposed addition of March and April 1 – 15 is finalized, revising season to March – October 15.

Wyoming: Proposed addition of January, February, March, and removal of October is finalized, revising season to January – September.

As discussed above, there is strong evidence supported in the literature that background O_3 across the U.S. is highest at many sites across the U.S. during the springtime (including into the month of June) and is an important contributor at many high-elevation sites throughout the year. As noted earlier (Section 3.1.2), actual O_3 monitoring data show that the highest O_3 exposures for the Park sites occur across the U.S. during the springtime and into early summer (i.e., March-June). As indicated earlier, 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 Parks with O_3 monitors for the period 2006-2010. Using hourly average O_3 data from 57 National Parks, Table 7A-2 shows that several of the O_3 monitors in the Parks experienced their highest W126 exposures during the spring months and early summer.

One site not included in the table summarizing the analysis of O_3 monitoring data for the Parks in the EPA's 2014 PA (EPA, 2014c) (Welfare Appendix, page 7A-12) was the Look Rock site (TN) (470090101) in the Great Smoky Mountain NP (GRSM). Table 3-4 below illustrates the top-10 daily maximum 8-h average concentrations and the date/time associated with each occurrence. Beginning in 1989 and continuing to the present, the Park began monitoring at Look Rock (823 m), located on the Foothills Parkway on the TN side of the Park. As mentioned earlier, the Park has historically been subject to elevated O_3 levels (Neufeld et al., 2019). 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. Data from the Look Rock O_3 monitor were used in their analyses. The highest hourly average concentration in the Park was recorded at the Look Rock site at 1600h on August 25, 1998. Note that in the early years (e.g., 1988), the top-10 8-h average concentrations occurred during the summer months. In 2018, all the top-10 8-

h values occurred during the March-June period. The Look Rock (TN) site exhibited a shift of its top-10 8-h values from the summer to the March-June period.

Table 3-4. Top-10 daily maximum 8-h average concentrations and the date/time associated with each occurrence for Look Rock (TN) (470090101) O_3 monitoring site in Great Smoky Mountains National Park. All available data over the entire period of record were included in this analysis independent of the EPA-defined O_3 season.

Top-10	1998		1999		2005		2009		2012		2018	
1st	0.122	8/25/1998 14:00	0.110	7/23/1999 18:00	0.092	4/19/2005 14:00	0.084	6/25/2009 13:00	0.078	6/29/2012 12:00	0.073	6/7/2018 16:00
2nd	0.116	9/12/1998 16:00	0.110	8/18/1999 10:00	0.089	9/12/2005 15:00	0.070	6/24/2009 16:00	0.077	6/27/2012 18:00	0.069	6/5/2018 15:00
3rd	0.112	9/11/1998 14:00	0.107	9/4/1999 12:00	0.086	4/17/2005 16:00	0.069	4/18/2009 12:00	0.077	6/28/2012 13:00	0.068	6/15/2018 16:00
4th	0.110	9/2/1998 14:00	0.106	8/17/1999 19:00	0.086	7/25/2005 14:00	0.068	4/9/2009 15:00	0.075	7/1/2012 14:00	0.067	5/11/2018 13:00
5th	0.104	8/24/1998 20:00	0.105	9/1/1999 12:00	0.085	4/18/2005 12:00	0.068	4/17/2009 15:00	0.073	6/30/2012 12:00	0.066	3/16/2018 13:00
6th	0.103	9/4/1998 13:00	0.104	8/4/1999 17:00	0.085	6/22/2005 14:00	0.066	3/23/2009 12:00	0.071	6/24/2012 14:00	0.066	4/21/2018 13:00
7th	0.102	9/5/1998 16:00	0.104	8/6/1999 13:00	0.085	6/25/2005 12:00	0.066	6/26/2009 19:00	0.071	8/2/2012 15:00	0.065	4/12/2018 16:00
8th	0.099	9/17/1998 13:00	0.102	7/26/1999 13:00	0.084	5/18/2005 17:00	0.065	8/13/2009 14:00	0.070	6/22/2012 13:00	0.065	5/1/2018 15:00
9th	0.098	8/22/1998 15:00	0.102	9/2/1999 15:00	0.084	9/9/2005 13:00	0.064	4/8/2009 12:00	0.069	4/14/2012 17:00	0.064	4/13/2018 11:00
10th	0.098	8/29/1998 12:00	0.101	9/3/1999 14:00	0.084	9/20/2005 15:00	0.064	6/2/2009 10:00	0.069	6/23/2012 15:00	0.064	5/12/2018 17:00

Source: EPA AQS data run.

Table 3-5. Top-10 daily maximum 8-h average concentrations and the date/time associated with each occurrence for Carter County (KY) (210430500) O_3 monitoring site. All available data over the entire period of record were included in this analysis independent of the EPA-defined O_3 season.

Top-10	1998		2002		2008		2018	
1st	0.104	9/13/1998 11:00	0.094	8/3/2002 10:00	0.082	4/18/2008 10:00	0.064	4/13/2018 10:00
2nd	0.100	8/22/1998 11:00	0.093	9/9/2002 10:00	0.078	8/19/2008 11:00	0.064	4/27/2018 11:00
3rd	0.097	9/12/1998 11:00	0.088	6/22/2002 11:00	0.075	7/16/2008 11:00	0.063	5/1/2018 11:00
4th	0.096	8/23/1998 10:00	0.086	9/10/2002 10:00	0.072	5/1/2008 11:00	0.063	5/11/2018 11:00
5th	0.090	9/6/1998 11:00	0.083	7/5/2002 10:00	0.071	4/17/2008 11:00	0.061	4/21/2018 11:00
6th	0.090	9/7/1998 11:00	0.083	9/8/2002 10:00	0.070	5/6/2008 11:00	0.059	4/17/2018 12:00
7th	0.089	8/7/1998 10:00	0.080	8/9/2002 11:00	0.070	8/20/2008 11:00	0.058	5/12/2018 10:00
8th	0.088	8/20/1998 11:00	0.080	9/7/2002 10:00	0.069	5/7/2008 10:00	0.057	4/18/2018 13:00
9th	0.087	9/14/1998 10:00	0.079	5/24/2002 11:00	0.069	8/22/2008 11:00	0.057	5/9/2018 10:00
10th	0.086	5/15/1998 11:00	0.079	8/13/2002 10:00	0.067	5/29/2008 11:00	0.057	5/10/2018 11:00

Source: EPA AQS data run.

In addition to the Tennessee example, Blanchard et al. (2019) noted that the highest peak 8-h O_3 maxima typically occurred in summer throughout New York state prior to about 2010. Annual maxima now occur during spring at rural locations but continue to persist in summer in the New York City metropolitan area. Similarly, in the southeastern U.S., Blanchard and Hidy (2019) reported that the highest peak daily 8-h average O_3 concentrations tended to occur in summer, but the authors provide some evidence for a recent shift in the frequency of maxima to spring in some locations.

There are many sites within the EPA's AQS database that show similar shifting monthly patterns for the top-10 8-h values from the summer to the March-June period. For example, a site in Carter County, KY (210430500) illustrates the shifting pattern from the summer to the March-April months for the 1998-2018 period. The site is listed in the AQS database as rural residential. Table 3-5 (please see the table above) illustrates the top-10 daily maximum 8-h average concentrations and the date/time associated with each occurrence. In addition, there are many other sites in the AQS database that illustrate their highest 8-h daily maximum concentrations during the spring. The EPA performed an analysis in 2014, which addressed whether extensions of currently required monitoring seasons were appropriate (Rice, 2014). Performing an additional analysis and placing the results into the draft Ozone National Ambient Air Quality Standards document would add clarity to the situation and provide an update to the Agency's 2014 analysis.

In summary, in this section we identified several sites with bell-shaped-like distribution curves of the hourly average O_3 concentrations. We saw for the Yellowstone NP (WY) site, a location that does not exhibit statistically significant trends for the MDA8 metric using the Mann-Kendell nonparametric test, that the most frequent hourly average O_3 concentrations were in the 40-45 ppb range. Fig. 3-49 illustrates the average relative contributions of current hourly background (blue) and anthropogenic O_3 (red) in 2006 (Lefohn et al., 2014). The black line shows the distribution pattern overlaid onto the figure. Note that the distribution pattern observed (black line) in 2006 in Fig. 3-49 resembles the distribution pattern illustrated in Fig. 3-44 for 2018 and there appears to be little influence of anthropogenic sources. Fig. 3-49 described by Lefohn et al. (2014) illustrates that the percent background O_3 concentrations in the 40-45 ppb compared to total observed O_3 values consist mostly of estimated background O_3 concentrations. For those sites that are more influenced by anthropogenic sources than the Yellowstone NP site, as emission reductions occur, a compression of the distribution of hourly average O_3 concentrations (with the higher values shifting downward toward the mid-values and the lower values shifting upward toward the mid concentrations) occurs. As mentioned earlier in this section, as emission reductions continue to decrease the absolute value of the higher concentrations, background O_3 increases its role as indicated by the percent contribution to the total O_3 measured. As U.S. anthropogenic contributions approach zero (which is not possible socially), what will remain is a distribution of hourly average O_3 concentrations that represent background O_3 that in many cases for inland sites is described as the bell-shaped-like curve. While we cannot say at what point the bell-shaped-like curve will no longer change except for meteorological variability, if the curve remains stable from year-to-year except for minor variability, then we have the ability to gain insight about the distribution of background O_3.

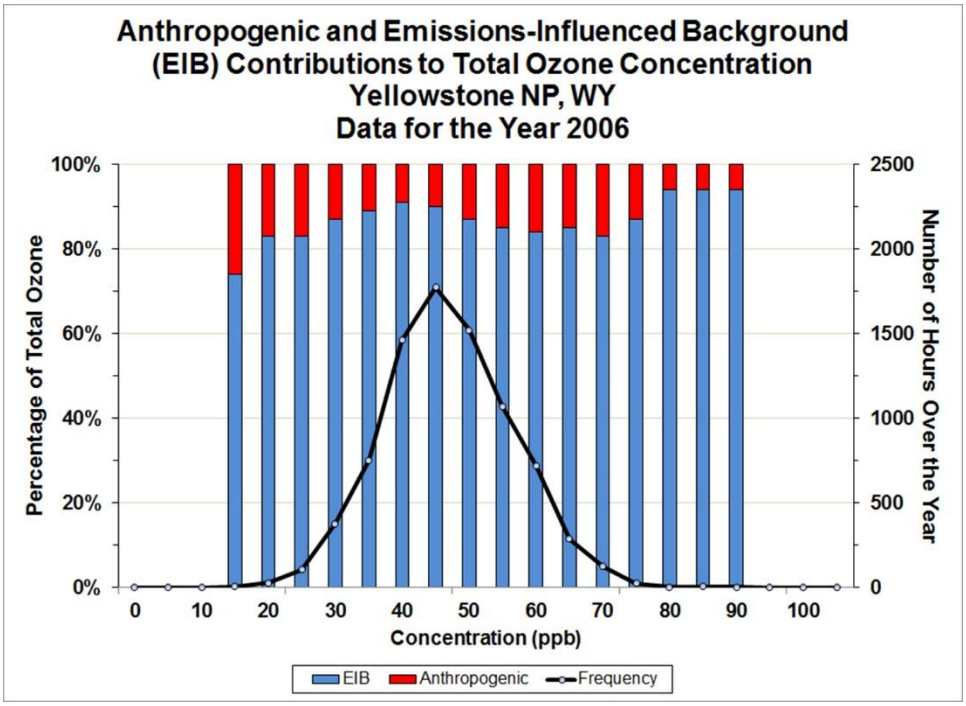


Figure 3-49. Average relative contributions of hourly background (blue) and anthropogenic O_3 (red) for Yellowstone NP (WY) (AQS ID 560391011) in 2006. The black line indicates the distribution of the hourly average O_3 concentrations. (Source: Lefohn et al., 2014).

Besides discussing the compression of the hourly average O_3 concentrations, we discussed a pattern where many sites within the EPA's AQS database show shifting from the summer months to the March-June period for the higher daily maximum 8-h values. This pattern is important. As will be discussed in the next section, the seasonal change pattern provides us with the ability to overlay this information with the predictions indicated by background O_3 modeling.

3.2.9 Observed Ozone Exposure Patterns and Model Performance

The USB modeling results described in the PA (EPA, 2020) indicate the following seasonal patterns:

- The current analysis indicates that natural and USA O_3 contributions peak during the traditional O_3 season (May through September), while long-range intercontinental transport of international O_3 (i.e. contributions from China, India etc.) peaks in the spring (February through May). (page 2-64).

- The natural contribution has a single maximum in late summer in the West, whereas, in the East there is evidence of two peaks— the largest in late Spring and a second peak in early Fall. (page 2-48).

As indicated in Section 3.2.4 and the previous section, the USB modeling results summarized in the first bullet above as indicated in the PA (EPA, 2020b) show a different seasonal pattern for when background O_3 is highest than the pattern reported in previous published results, including the EPA's own reports (EPA, 2013, 2014a). The bullets indicate that the Natural component (which is the largest contributor to background O_3) of the current EPA model is predicted to occur in the West in late summer. The previous conclusion in the 2014 PA (EPA, 2014a) and the 2013 ISA (EPA, 2013, in section 3.4) was that background O_3 was greatest over the U.S. during the spring and early summer (i.e., June).

Bias adjustment to estimated background O_3 estimates has been described in the literature. Dolwick et al. (2015) and Lefohn et al. (2014) used bias-adjusted estimates for USB_{AB} and EIB, respectively. Lefohn et al. (2014) concluded that, based on the tendency for their model to underestimate STT processes, the upward adjusted values of the hourly EIB concentration range were preferable to using an average or median value of the hourly range, especially during the spring at high-elevation sites. In their analyses, Lefohn et al. (2014) noted that model performance at low-elevation sites tended toward larger under prediction biases in cool months (i.e., November-April) and larger over prediction biases in warm months (June-October), particularly for sites in the southern and eastern U.S. The authors attributed site-specific monthly under and over predictions to their Global background O_3 (GBO_3) or anthropogenic O_3 modeling components. Global background O_3 (GBO_3) was defined as the sum of the global tropospheric and stratospheric components. For almost all high-elevation sites in their analyses, the model under predicted in the spring months when the above-median MDA8 GBO_3 coincided with diagnosed STT-S events as per the stratospheric trajectory analyses performed by Dr. Heini Wernli, Institute for Atmospheric and Climate Science, ETH Zurich in Switzerland. Table 3-6 (Table 4 from Lefohn et al., 2014), summarizes the months when STT-S events coincided with months of highest MDA8 GBO_3 and the model tended to be under (u) or over (o) predicted. As indicated above, for almost all high-elevation sites, the model under predicted in the spring when the above-median MDA8 GBO_3 coincided with diagnosed STT-S events. Given the relatively small spring anthropogenic contributions at these sites, Lefohn et al. (2014) believed it was likely that the underestimates were associated with GBO_3. For some urban sites (i.e., Boston, Dallas, Detroit, New York, and Sacramento), the model also under predicted during the spring when higher GBO_3 coincided with diagnosed STT-S events; for others (i.e., Atlanta, Baltimore, Chicago, Cleveland, Georgia Station, Houston, Philadelphia, St. Louis, and Washington DC), spring over predictions occurred when higher GBO_3 coincided with STT-S events. For urban sites with higher spring anthropogenic influences, it was more likely that over predictions were associated with anthropogenic O_3.

The important role that the stratosphere played in episodic (i.e., short-term, high concentration events), as well as enhancements (subtle increases in O_3 concentration), to surface O_3 values was noted in Lefohn et al. (2012). The authors quantified the frequency of stratosphere-troposphere exchange (STE) 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 U.S. during the years 2007-2009. The authors employed a forward trajectory-based approach to address the relationship between stratospheric intrusions and enhancements in hourly average O_3 concentrations. The authors results indicated that STT down to the surface (STT-S) frequently contributed to enhanced surface O_3 hourly averaged concentrations at sites across the U.S., 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 U.S. experienced enhanced O_3 concentrations coincident with STT-S events. Tables 3-7 – 3-12 (Supplemental Tables S-1 – S-6 in Lefohn et al., 2012) provide a detailed description of the number of days in which the daily maximum hourly average O_3 concentration was $\geq$ 50 ppb and coincident with a direct STT-S event for each of the statistically significant STT-S months. The tables provide an indication of the variability of the coincidences at each site across years.

Table 3-6. Months when STT-S events coincided with months of highest MDA8 GBO_3 and the model tended to be under (u) or over (o) predicted. Source: Reproduced from Table 4 in Lefohn et al. (2014).

Site Name	Jan	Feb	Mar	Apr	May	June	July	Aug	Sept	Oct	Nov	Dec
High Elevation												
Yosemite NP, CA	o	o	u	u	u	u		u		u		
Denver, CO	u	u	u	u	u	u						
Gothic, CO	u	o	u	u	o	o						
Pinedale, WY		u	u	u	u							
Yellowstone NP, WY		u	u	u	u	u	o	o				
Shenandoah NP, VA		o	o	u								
Low Elevation												
Atlanta, GA			o	o								
Baltimore, MD			o									
Boston, MA		u										
Chicago, IL		o										
Cleveland, OH				o								
Dallas, TX			u									
Detroit, MI					u							
Georgia Station, GA				o								
Houston, TX			o									
Los Angeles, CA		o		o	u	u						
New York, NY				u								
Philadelphia, PA					o							
Sacramento, CA		o		u	u							
Seattle, WA												
St. Louis, MO				u	o					o		
Voyageurs NP, MN					u							
Washington DC			o									

On page 2-66 of the PA (EPA, 2020b), the authors note

> For this analysis, we did not attempt to quantify the contributions from individual Natural sources (e.g., lightning, soil, fires, stratosphere) or to address exceptional events beyond basic screening to remove very large fire plumes.

On page 2-41 of the PA (EPA, 2020b), the authors note

> Near the tropopause, there is a low bias in the model that is most pronounced in the spring. The low bias at the tropopause likely suggests an underestimate of stratospheric exchange. Mean bias drops to below 20% in the middle troposphere (600-300 hPa). The low-bias in the free troposphere may stem from underestimation of spring time stratospheric contribution in some regions.

On page 2-42 of the PA (EPA, 2020b), the authors also note

> Dolwick et al., 2015) showed that multi-model estimates converged when applying bias correction, indicating that differences in USB estimates are correlated with model performance. No bias correction has been applied here, so in a limited manner bias in ambient predictions can help set expectations for bias in USB. Based on hemispheric model evaluation, the stratospheric component in spring is likely underestimated leading to a USB low bias in spring.

Not applying a bias correction to the estimated USB concentrations when observed biases in the model were observed may explain why the seasonal patterns exhibited in the model described in the PA (EPA, 2020b) did not match the patterns observed when one characterizes ambient data (see previous section). As noted above, the EPA model described in the PA (EPA, 2020b) showed biases in the springtime. As noted in Tables 3-7 – 3-12 (Supplemental Tables S-1 – S-6 in Lefohn et al., 2012), the spring months are when the stratosphere plays an important role at both low- and high-elevation O_3 surface sites across the U.S. Enhanced, as well as episodic, contributions to ambient O_3 levels occur from the stratosphere during this time period. The USB low bias in spring may explain some of the inconsistency observed in the seasonal patterns for USB observed in the EPA modeling results described in the PA (EPA, 2020b).

Table 3-7. Months in which highest-elevation (> 2.3 km) monitoring sites in the Intermountain West exhibited a statistically significant coincidence value and there was a coincidence between the number of days with daily maximum hourly average O_3 concentrations ≥ 50 ppb and STT-S >0. The number of days during the specific month when the daily maximum hourly average concentration was ≥ 50 ppb and the STT-S was > 0 is in parenthesis (). Source: Table S-1 from Lefohn et al. (2012).

Site	Months
Yellowstone NP, WY	March 2007 (21), April 2007 (27), May 2007 (27), June 2007 (21), August 2007 (20), March 2008* (23), April 2008 (29), May 2008 (22), June 2008 (19), July 2008 (22), March 2009 (22), April 2009 (23), May 2009 (28), June 2009 (14)
Pinedale, WY	March 2007 (22), April 2007 (28), May 2007 (25), June 2007 (24), August 2007 (26), September 2007 (24), February 2008 (22), March 2008 (29), April 2008 (29), May 2008 (20), June 2008* (17), July 2008 (25), August 2008* (15), September 2008* (13), March 2009 (30), April 2009 (26), May 2009 (23), July 2009 (22)
Centennial, WY	April 2007 (25), May 2007 (20), June 2007 (25), August 2007 (20), September 2007 (24), February 2008 (21), March 2008 (30), April 2008 (30), May 2008 (28), June 2008 (25), July 2008 (24), August 2008 (25), September 2008 (20), March 2009 (29), April 2009 (27), May 2009* (9), June 2009* (17), July 2009 (19), August 2009 (23), September 2009 (24)
Gothic, CO	March 2007 (24), April 2007 (28), May 2007 (27), June 2007 (21), September 2007 (21), February 2008 (22), March 2008 (21), April 2008* (11), May 2008* (22), July 2008 (24), February 2009 (21), March 2009 (28), April 2009 (30), May 2009 (24), June 2009 (20), July 2009 (21), August 2009 (21)
Rocky Mountain NP, CO	March 2007 (23), April 2007 (23), May 2007 (23), June 2007 (20), September 2007 (25), February 2008 (20), March 2008 (29), April 2008 (30), May 2008 (26), June 2008 (27), July 2008 (25), August 2008 (20), September 2008 (19), March 2009 (29), April 2009 (25), May 2009 (24), June 2009 (23), July 2009 (19), August 2009 (20), September 2009 (21), November 2009 (18), December 2009 (23)

*Data capture less than 90% but statistically significant coincidences existed.

Table 3-8. Months in which higher-elevation (1.5 – 2.2 km) monitoring sites in the Intermountain West exhibited a statistically significant coincidence value and there was a coincidence between the number of days with daily maximum hourly average O_3 concentrations ≥ 50 ppb and STT-S >0. The number of days during the specific month when the daily maximum hourly average concentration was ≥ 50 ppb and the STT-S was > 0 is in parenthesis (). Source: Table S-2 from Lefohn et al. (2012).

Site	Months
Mesa Verde NP, CO	March 2007 (23), April 2007 (28), May 2007 (22), September 2007 (19), October 2007 (18), March 2008 (28), April 2008 (29), May 2008 (27), June 2008 (21), September 2008 (14), March 2009 (25), April 2009 (28), May 2009 (24), June 2009 (25), September 2009 (18)
Grand Canyon NP, AZ	February 2007 (13), March 2007 (23), April 2007 (26), May 2007 (27), September 2007 (25), October 2007 (18), February 2008 (20), March 2008 (29), April 2008 (29), May 2008 (28), June 2008 (20), September 2008 (18), March 2009 (21), April 2009 (26), May 2009 (24), June 2009 (26), September 2009 (16)
Canyonlands NP, UT	April 2007 (29), May 2007 (25), June (19), September 2007 (23), March 2008 (28), April 2008 (29), May 2008 (28), June 2008 (25), July 2008 (20), August 2008 (22), September 2008 (21), February 2009 (14), March 2009 (27), April 2009 (28), May 2009 (26), June 2009 (24), July 2009 (21), August 2009 (20), November 2009 (2)
Chiricahua NM, AZ	March 2007 (24), April 2007 (29), May 2007 (19), October 2007 (12), February 2008 (18), March 2008 (27), April 2008 (29), May 2008 (27), October 2008 (16), February 2009 (17), March 2009 (24), April 2009 (30), May 2009 (25)
Great Basin NP, NV	April 2007 (25), May 2007 (31), June 2007 (24), August 2007 (21), September 2007 (23), March 2008 (25), April 2008 (27), May 2008 (26), June 2008 (25), July 2008 (21), August 2008 (21), March 2009 (26), April 2009 (29), May 2009 (26), June 2009* (23), September 2009 (18)
Jefferson County, CO	March 2007 (21), April 2007 (16), May 2007 (21), September 2007 (22), March 2008 (27), April 2008 (29), May 2008 (25), June 2008 (24), July 2008 (20), March 2009 (24), April 2009* (19), May 2009 (20), June 2009 (20), September 2009 (18)

*Data capture less than 90% but statistically significant coincidences existed.

Table 3-9. Months in which high-elevation (> 1.3 km) monitoring sites in the West and East exhibited a statistically significant coincidence value and there was a coincidence between the number of days with daily maximum hourly average O_3 concentrations ≥ 50 ppb and STT-S >0. The number of days during the specific month when the daily maximum hourly average concentration was ≥ 50 ppb and the STT-S was > 0 is in parenthesis (). Source: Table S-3 from Lefohn et al. (2012).

Site	Months
Lassen Volcanic NP, CA	March 2007 (21), April 2007 (23), May 2007 (29), June 2007 (21), July 2007 (19), August 2007 (24), September 2007 (22), March 2008 (29), April 2008 (29), May 2008 (20), June 2008 (26), July 2008 (30), August 2008 (25), September 2008 (24), March 2009 (24), April 2009 (24), May 2009 (23), July 2009 (22), August 2009 (21), September 2009 (19)
Yosemite NP, CA (Turtleback Dome)	April 2007 (27), May 2007 (29), June 2007 (23), August 2007 (22), September 2007 (25), October 2007 (20), March 2008 (27), April 2008 (29), May 2008 (26), June 2008 (24), July 2008* (22), August 2008* (17), September 2008 (26), October 2008 (23), March 2009 (24), April 2009 (27), May 2009 (24), June 2009 (16), July 2009 (28), August 2009 (22), September 2009 (24)
Crestline, CA	March 2007 (26), April 2007 (24), May 2007 (26), October 2007 (20), March 2008 (21), April 2008 (28), May 2008 (29), June 2008 (21), September 2008 (24), October 28 (22), March 2009 (20), April 2009 (27), May 2009 (24), June 2009 (17), September 2009 (19)
Mount Washington, NH	March 2007 (17), May 2007 (20), September 2007 (15), March 2008 (19), April 2008 (23), May 2008 (23), June 2008 (16), March 2009 (25), April 2009 (21), May 2009 (24), September 2009 (15), November* 2009 (10)
Whiteface Mountain, NY	March 2007 (17), May 2007 (21), March 2008 (20), April 2008 (22), May 2008 (20), April 2009 (16), May 2009 (22), September 2009 (9)

*Data capture less than 90% but statistically significant coincidences existed.

Table 3-10. Months in which low-elevation (< 1.3 km) monitoring sites in the West and Intermountain West exhibited a statistically significant coincidence value and there was a coincidence between the number of days with daily maximum hourly average O_3 concentrations ≥ 50 ppb and STT-S >0. The number of days during the specific month when the daily maximum hourly average concentration was ≥ 50 ppb and the STT-S was > 0 is in parenthesis (). Source: Table S-4 from Lefohn et al. (2012).

Site	Months
Cheeka Peak, WA	May 2007 (7), August 2007 (1), June 2009 (3), September 2009 (1)
King County, WA	January 2007 (1), August 2008 (1), July 2009 (1)
Mount Rainier NP, WA	August 2008 (4), July 2009 (9)
Trinidad Head, CA	June 2008 (2)
El Dorado County, CA	May 2007* (11), September 2007 (18), March 2008 (23), April 2008 (28), May 2008 (23), June 2008 (21), October (15), April 2009 (22), June 2009 (17), September 2009 (20)
Glacier NP, MT	April 2008 (18), April 2009 (18)
Big Bend NP, TX	February 2007 (13), March 2007 (25), April 2007 (22), September 2007 (6), October 2007 (11), March 2008 (25), April 2008 (28), August 2008 (5), September 2008 (6), February 2009 (17), March 2009 (16), April 2009 (28), June 2009 (7)

*Data capture less than 90% but statistically significant coincidences existed.

Table 3-11. Months in which low-elevation (< 1.3 km) monitoring sites in the Midwest exhibited a statistically significant coincidence value and there was a coincidence between the number of days with daily maximum hourly average O_3 concentrations ≥ 50 ppb and STT-S >0. The number of days during the specific month when the daily maximum hourly average concentration was ≥ 50 ppb and the STT-S was > 0 is in parenthesis (). Source: Table S-5 from Lefohn et al. (2012).

Site	Months
Theodore Roosevelt NP, ND	April 2007 (16), May 2007 (15), April 2008 (22), May 2008 (20), May 2009 (17)
Voyageurs NP, MN	April 2007 (14), September 2007 (4), April 2008 (18), April 2009 (19), May 2009 (18)
Ann Arbor, MI	April 2007 (15), May 2007 (21), June 2007 (17), April 2008 (20), May 2008 (22), July 2008 (16), May 2009 (23), June 2009 (18)
Cook County, IL	June 2007 (14), April 2008 (15), June 2008 (15), April 2009 (14), May 2009 (17), July 2009 (13)
Alhambra, IL	April 2007 (22), June 2007 (16), July 2007 (17), April 2008 (19), May 2008 (22), June 2008 (17), April 2009 (14), May 2009 (16), September 2009 (8)
Stockton, IL	August 2009* (1)
Harris County, TX	March 2007 (6), May 2007 (9), June 2007 (1), April 2008 (12), May 2008 (10), June 2008 (1), July 2008 (3), September 2008* (4), October 2008 (16), March 2009 (11), April 2009 (13), May 2009 (5), June 2009 (4), July 2009 (1), August 2009 (6), September 2009 (4)

*Data capture less than 90% but statistically significant coincidences existed.

Table 3-12. Months in which low-elevation (< 1.3 km) monitoring sites in the East exhibited a statistically significant coincidence value and there was a coincidence between the number of days with daily maximum hourly average O_3 concentrations ≥ 50 ppb and STT-S >0. The number of days during the specific month when the daily maximum hourly average concentration was ≥ 50 ppb and the STT-S was > 0 is in parenthesis (). Source: Table S-6 from Lefohn et al. (2012).

Site	Months
Georgia Station, GA	March 2007 (22), April 2007 (23), May 2007 (20), June 2007 (19), July 2007 (8), September 2007 (13), March 2008 (23), April 2008 (20), May 2008 (23), July 2008 (15), March 2009 (14), April 2009 (18), May 2009 (6)
Rockdale, GA	March 2007 (21), April 2007 (23), May 2007* (17), March 2008 (21), April 2008 (18), May 2008 (25), September 2008 (8), May 2009 (8), July 2009 (14)
Cuyahoga County, OH	April 2008 (19), May 2008 (20), May 2009 (16) July 2009 (15)
Bucks County, PA	May 2007 (19), June 2007 (17), July 2007* (13), May 2008 (16), April 2009 (15), May 2009 (16), June 2009 (10)
Shenandoah NP, VA	April 2007 (19), May 2007 (22), June 2007* (18), September 2007 (21), April 2008 (22), May 2008 (27), June 2008 (18), July 2008 (18), August 2008 (20), September 2008 (10), March 2009 (16), April 2009 (21), May 2009 (13), June 2009 (14), August 2009 (10)
Blackwater NWR, MD	September 2008 (10), May 2009 (10)
Abington, CT	July 2008 (14), April 2009 (14)
Fairfield, CT	April 2009 (15), June 2009 (11)
Chittenden County, VT	April 2008 (19), May 2008 (17), April 2009 (18), May 2009 (17)

*Data capture less than 90% but statistically significant coincidences existed.

Jaffe et al. (2018) discussed a site in Colorado, where simulations and contributions were compared for a monitor at Chatfield (AQS 08-035-0004) for May-September 2011. The authors described the site as a regulatory relevant suburban monitor southwest of Denver, Colorado. Figure 6 in Jaffe et al. (2018) showed the observed and modeled daily MDA8 values using results from an EPA model. Monthly averaged biases at the Chatfield monitor were marginally negative in the EPA simulations. The authors noted that Fig. 6 in Jaffe et al. (2018) suggested four distinct segments of performance and simulated contributions at the Chatfield monitor that were related to contributions from noncontrollable O_3 sources. As pointed out by the authors, the simulations started in a USB O_3 dominated regime (May 1 to June 7), then went through a transition period (June 8 to July 15), and then ended with two periods dominated by local contributions (July 16 to August 22 and August 23 to September 29). During the USB O_3 dominated period, the EPA model experienced a mean bias of -2.1. During the transition period, the EPA simulations performed poorly and experienced a mean bias of -3.6. During the locally dominated period of July 16 to August 22, the simulation performed well with a mean bias of -0.9. During the locally dominated period of August 23 to September 29, the mean bias was -3.3. Jaffe et al. (2018) noted that the simulations performed better during periods of sustained contribution (USB O_3 or local). The authors noted that simulations performed even better when USB O_3 and local contribution were not anti-correlated, and simulations performed best when local contributions were dominant. The model performed well for average biases, but model correlation with observations was better when local contributions were dominant and when anti-correlation between local and USB O_3 contributions was weak.

Fig. 3-50 illustrates for the Chatfield site in 2011 the comparison of observed daily O_3 MDA8 concentrations and the STT-S counts (estimated for a site in the Denver area for 2011). The USB O_3 dominated regime identified in Jaffe et al. (2018) (May 1 to June 7) was a period of large numbers of STT-S counts, the transition period (June 8 to July 15) exhibited a declining number of STT-S counts, the period July 16 to August 22 experienced fewer STT-S counts, and the period August 23 to September 29 exhibited an increasing pattern of STT-S counts. The period from early July to early September was the time when the lowest number of STT-S counts occurred over the 2011 period. Based on the mean monthly bias observed by Jaffe et al. (2018) for the high-elevation site at Chatfield, the periods associated with greatest biases in the EPA model appeared to be associated with the period when the contribution of STT-S was most important. The lowest mean bias was -0.9 which occurred during July 16 to August 22, a period when the STT-S counts were the lowest.

The PA (EPA, 2020b, page 2-42) indicates, based on hemispheric model evaluation, that the stratospheric component in spring was likely underestimated leading to a USB low bias in spring. The authors noted that no bias correction had been applied to the EPA model used in the PA (EPA, 2020b). The PA (EPA, 2020b) observed that the Natural contribution has two peaks in the East. The largest occurred in late Spring and the second peak occurred in early Fall (EPA, 2020b, page 2-48). It is interesting to note that the pattern described in the PA (EPA, 2020b) for the East, was observed in both the East and West by Lefohn et al. (2012). The authors noted that for the 39 sites analyzed in their study, many of the sites during the spring, fall, and winter months, experienced higher GBO_3, which was associated with more frequent stratosphere-to-troposphere transport to the surface (STT-S) enhancements according to their independent three-dimensional trajectories based on global meteorological analyses. Patterns of higher spring

(Emissions Influenced Background (EIB) O_3 were followed by lower values during the summer, due to heightened chemical interaction with anthropogenic sources, which were then followed by rising EIB O_3 during the fall and winter months. For some high-elevation western U.S. sites, the seasonal pattern was less discernible due to relatively small anthropogenic contributions and the high EIB O_3 estimated throughout the year. EIB O_3 at all high-elevation sites contributed a significant proportion to total O_3 throughout the year and throughout the observed total O_3 frequency distribution, while EIB O_3 at most urban sites contributed a major portion to total O_3 during non-summer months and to the mid-range concentrations (30-50 ppb) of the frequency distribution. The different patterns in the West noted in EPA's USB model described in the PA (EPA, 2020b) and Lefohn et al. (2012) may be attributable to the lack of bias adjustment in the EPA model described in the PA (EPA, 2020b). One might hypothesize that if a bias adjustment were performed on the daily MDA8 predictions in the EPA model described in the PA (EPA, 2020b) that higher MDA8 values might have occurred during the spring and fall periods. If EPA had performed a bias adjustment, perhaps the EPA's USB modeling results might have agreed better with the seasonal USB patterns described in the Agency's 2014 PA (EPA, 2014a), as well as past publications in the peer-reviewed literature.

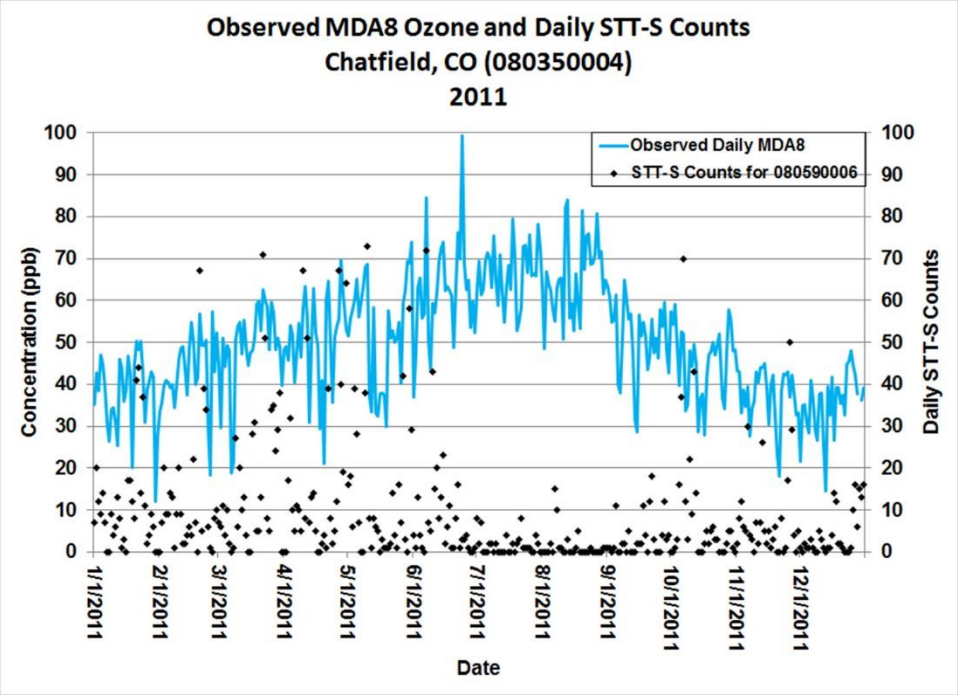


Figure 3-50. Observed MDA8 O_3 and daily STT-S counts for 2011 for the Chatfield (CO) (080350004) monitoring site. The STT-S counts, which were quantified for a site in Jefferson County (CO) (080590006), were superimposed over the observed data at the Chatfield site.

The PA (EPA, 2020b) does not provide examples for specific sites that describe the observed and USB concentrations for the eight sites used in the PA risk assessment (Atlanta, Boston, Dallas, Detroit, Philadelphia, Phoenix, Sacramento, and St. Louis). In the PA (EPA, 2020b), analyses are presented that estimate exposure and risk for simulated populations in eight study areas. The eight study areas represent a variety of circumstances about population exposure

to short-term concentrations of O_3 in ambient air. The eight study areas range in total population size from approximately two to eight million and are distributed across the U.S. in seven different NOAA climate regions: The Northeast, Southeast, Central, East North Central, South, Southwest and West. In Figs. 3-51 through 3-57, total observed O_3 concentrations, USB_{AB} estimates (USB_{AB} data provided by the EPA), and STT-S counts are presented for 2007 for seven of the eight sites (i.e., Atlanta, Boston, Dallas, Detroit, Philadelphia, Sacramento, and St. Louis) used by the EPA in its risk analyses presented in the PA (EPA, 2020b).

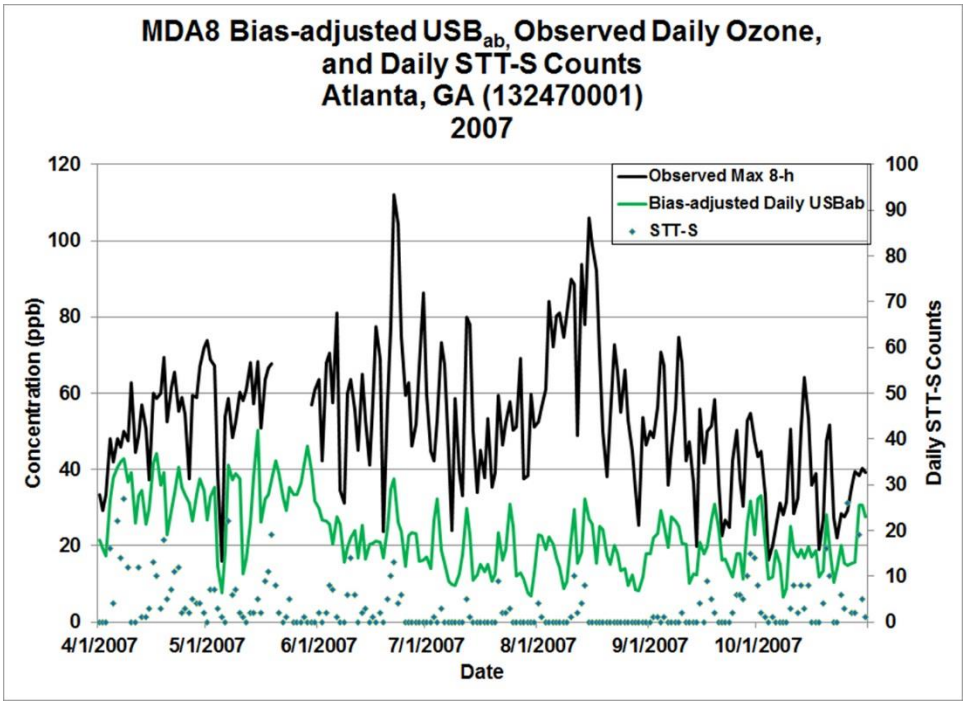


Figure 3-51. A comparison of the observed 8-h daily maximum concentration with the estimated bias-adjusted U.S. Background (USB_{AB}) 8-h daily maximum concentrations for a site in Atlanta, Georgia (AQS ID 132470001) for April-October 2007. The daily stratospheric-tropospheric transport to surface (STT-S) 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.

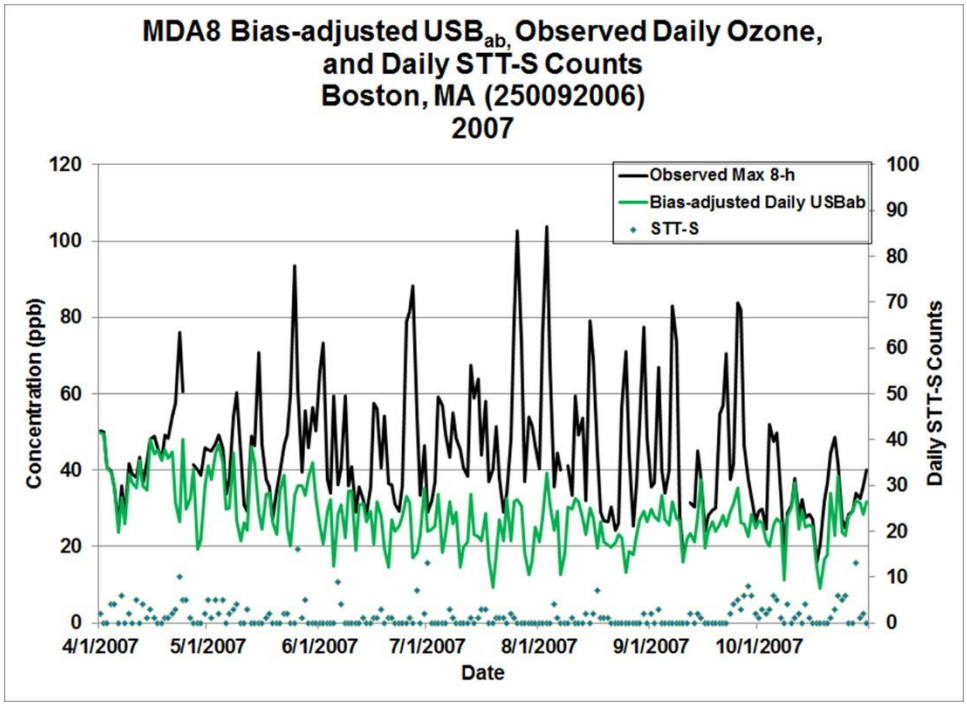


Figure 3-52. A comparison of the observed 8-h daily maximum concentration with the estimated bias-adjusted U.S. Background (USB_{AB}) 8-h daily maximum concentrations for a site in Boston, Massachusetts (AQS ID 250092006) for April-October 2007. The daily stratospheric-tropospheric transport to surface (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.

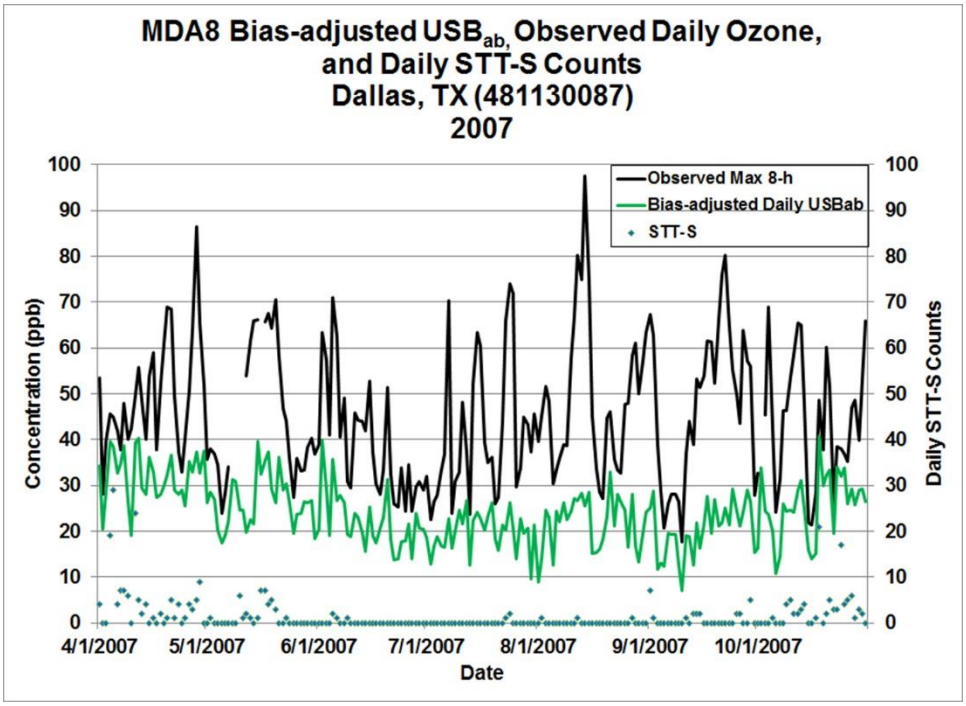


Figure 3-53. A comparison of the observed 8-h daily maximum concentration with the estimated bias-adjusted U.S. Background (USB_{AB}) 8-h daily maximum concentrations for a site in Dallas, Texas (AQS ID 481130087) for April-October 2007. The daily stratospheric-tropospheric transport to surface (STT-S) 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.

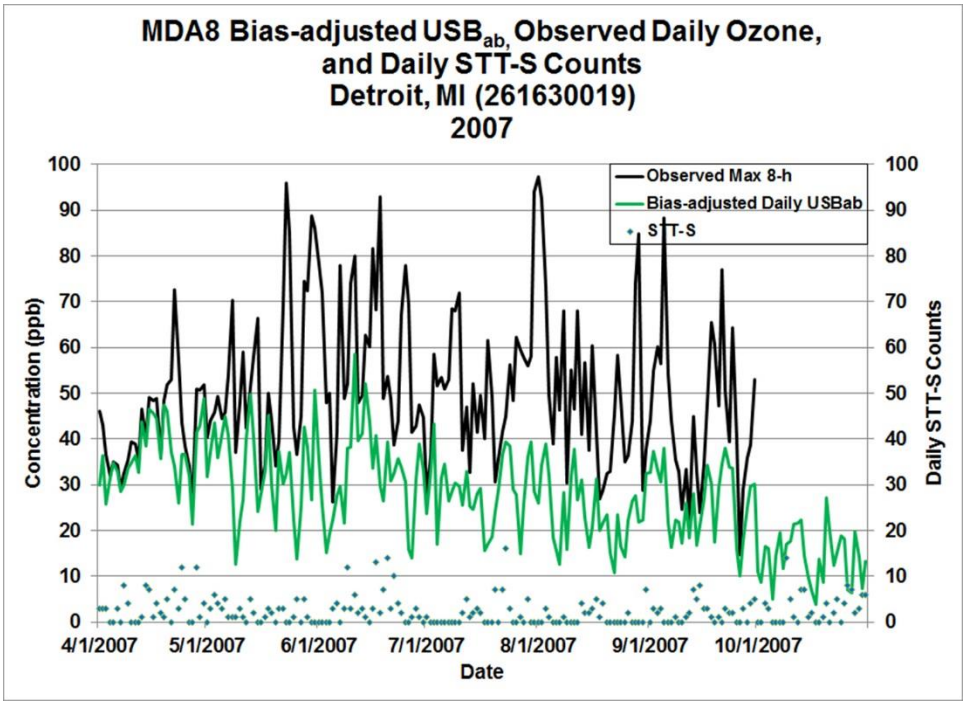


Figure 3-54. A comparison of the observed 8-h daily maximum concentration with the estimated bias-adjusted U.S. Background (USB_{AB}) 8-h daily maximum concentrations for a site in Detroit, Michigan (AQS ID 261630019) for April-October 2007. The daily stratospheric-tropospheric transport to surface (STT-S) 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.

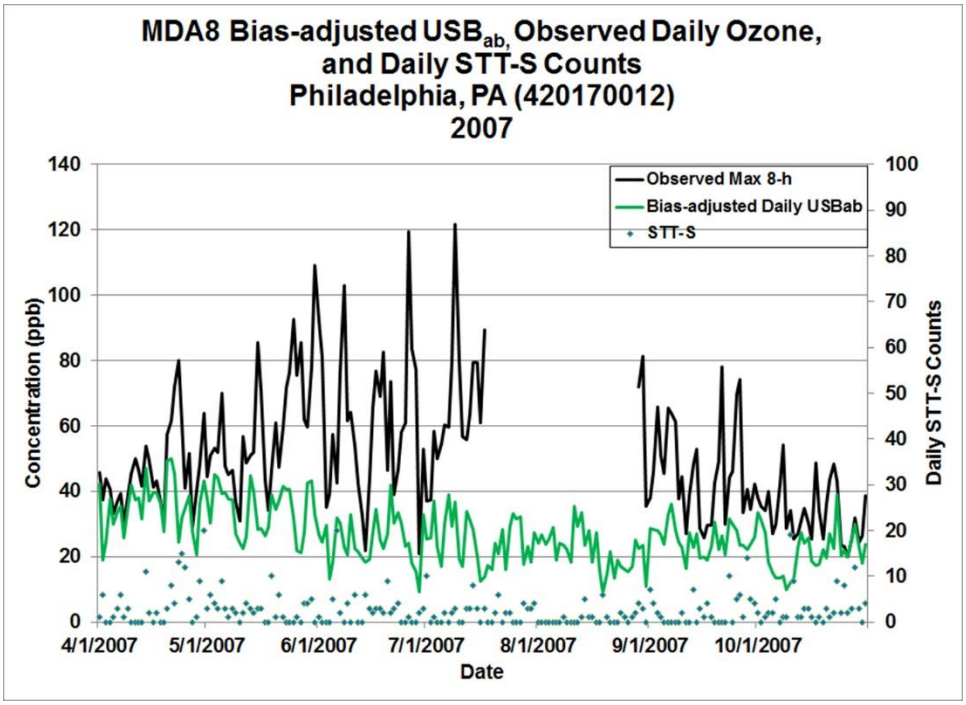


Figure 3-55. A comparison of the observed 8-h daily maximum concentration with the estimated bias-adjusted U.S. Background (USB_{AB}) 8-h daily maximum concentrations for a site in Philadelphia, Pennsylvania (AQS ID 420170012) for April-October 2007. The daily stratospheric-tropospheric transport to 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.

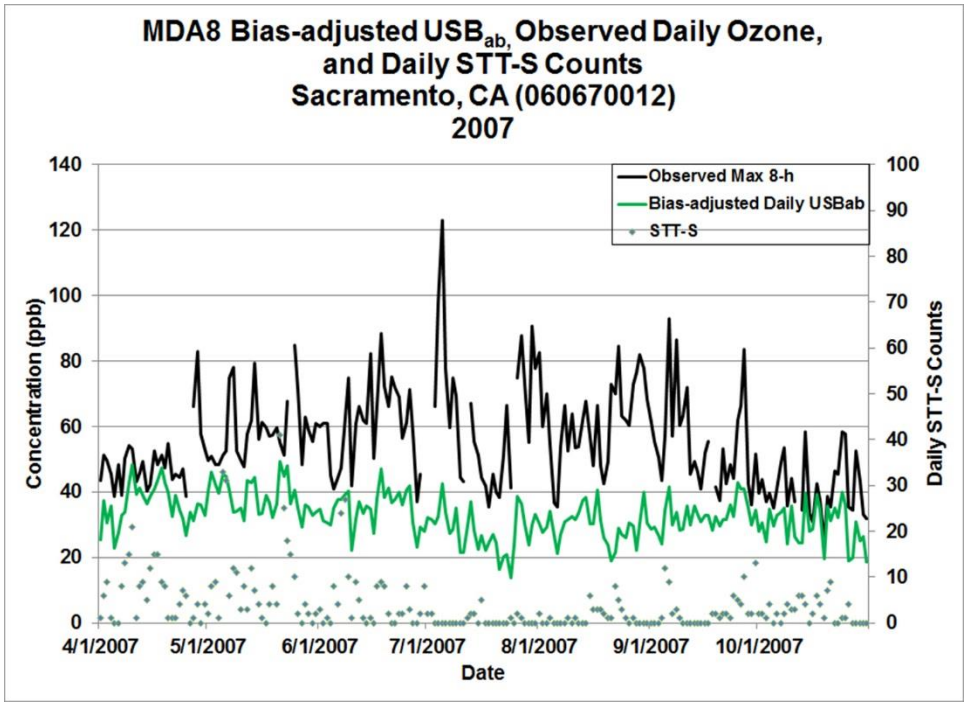


Figure 3-56. A comparison of the observed 8-h daily maximum concentration with the estimated bias-adjusted U.S. Background (USB_{AB}) 8-h daily maximum concentrations for a site in Sacramento, California (AQS ID 060670012) for April-October 2007. The daily stratospheric-tropospheric transport to surface (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.

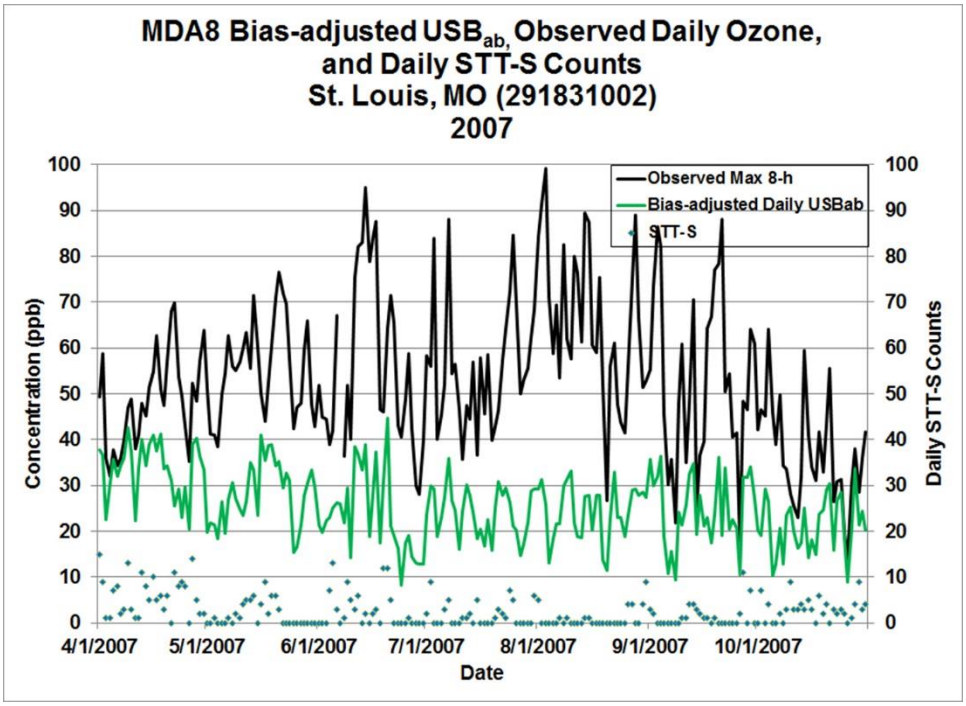


Figure 3-57. A comparison of the observed 8-h daily maximum concentration with the estimated bias-adjusted U.S. Background (USB_{AB}) 8-h daily maximum concentrations for a site in St. Louis, Missouri (AQS ID 291831002) for April-October 2007. The daily stratospheric-tropospheric transport to surface (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.

For many of the sites (Figs. 51-57), higher spring USB_{AB} O_3 was followed by lower values during the summer, which was then followed by rising USB_{AB} O_3 during the fall months. Focusing on the difference between the observed and USB_{AB} concentrations (i.e., **the Gaps**), the pattern varies by site. The Gaps indicate the apparent influence of anthropogenic sources on each site. The pattern of the difference between the observed and USB_{AB} concentrations (i.e., gaps) indicate for many of the seven sites the variability of USB_{AB} during specific periods. Enhanced O_3 levels associated with stratospheric contribution occur across the seven sites with the result that USB_{AB} contributes in varying amounts (i.e., depending upon season and location of the site) to the total observed O_3 concentrations across the U.S.

3.2.10 Model Performance USB versus USB_{AB}

In a perfect world when models perform well, the estimates of USB background O_3 should be higher than USB_{AB}. This is because the USB estimates are defined as the O_3 concentration that would occur if all U.S. anthropogenic O_3 precursor emissions were removed (ISA, 2020a, Page ES-3). While USB is an estimate of O_3 concentrations that could be achieved if all U.S. anthropogenic sources were eliminated, USB_{AB} is an estimate of how much O_3 can be attributed to background sources when those anthropogenic sources are still present. Background

O_3 is titrated by the NO associated with NO_x sources. Thus, it would be anticipated that USB estimated concentrations would generally be higher than the estimated USB_{AB} values, which represent titrated background O_3. Fig. 3-58 illustrates the difference between the estimates of USB and USB_{AB}, before and after a bias adjustment is made (Dolwick et al., 2015). Prior to the application of the bias-adjustment, there was a clear tendency for the CMAQ zero-out modeling to estimate higher levels of USB MDA8 O_3 concentrations compared to source apportionment USB_{AB} over most sites in the western U.S. As noted by the authors, constraining the USB and USB_{AB} estimates for model bias brings the estimates across the two methodologies closer together.

Fig. 3-59 illustrates the bias-adjusted model estimates for USB and USB_{AB} from the CMAQ and CAMx models, respectively (Dolwick et al., 2015). The spatial patterns for the April-October mean bias-adjusted MDA8 values between the two sets of estimates are similar. However, differences exist as one compares the site estimates using the two models. The differences in the estimates described in Dolwick et al. (2015) were based on 7-month mean bias-adjusted MDA8 values. If one examined the daily time series for USB and USB_{AB} MDA8 values, the differences in the estimates for USB and USB_{AB} would be greater. These daily differences are not quantified here because a comparison of daily USB and USB_{AB} values was not available to assess.

Dolwick et al. (2015) noted that because the two distinct model approaches estimated similar background impacts over the rural portions of the western U.S., the authors believed greater confidence could be placed on the combined results. However, the authors noted that while the CAMx and CMAQ model simulations provided consistent estimates in their study of rural USB O_3 levels in the western U.S., the CAMx source-apportionment approach (i.e., USB_{AB}) predicted lower background contributions in the urban areas than USB, as anticipated, because anthropogenic emissions reacted with and destroyed some fraction of the O_3 in the CAMx tracer species used to track the background O_3 contribution.

As noted in earlier sections, the USB modeling results described in the PA (EPA, 2020b) indicate the following seasonal patterns: (1) natural and USA O_3 contributions peak during the traditional O_3 season (May through September), while long-range intercontinental transport of international O_3 (i.e. contributions from China, India etc.) peaks in the spring (February through May) (page 2-64); (2) the natural contribution has a single maximum in late summer in the West, whereas, in the East there is evidence of two peaks— the largest in late Spring and a second peak in early Fall. (page 2-48). The previous conclusion in the 2014 PA (EPA, 2014a) and the 2013 ISA (EPA, 2013, in section 3.4) was that background O_3 was greatest over the U.S. during the spring and early summer (i.e., March-June period). While the estimates of the absolute values for background O_3 might differ depending upon the choice of whether USB or USB_{AB} was used in the modeling effort, I do not believe that the choice of USB or USB_{AB} explains the differences in the seasonal patterns associated with the background O_3 modeling results in the PA (EPA, 2020b) from previous reported patterns for background O_3.

As noted earlier, Dolwick et al. (2015) and Lefohn et al. (2014) used bias-adjusted estimates for USB_{AB} and EIB, respectively. The different pattern in the West noted in the PA (EPA, 2020b) compared to the pattern reported by Dolwick et al. (2015) and Lefohn et al. (2014)

may be attributable to the lack of bias adjustment in the current EPA USB model described in the PA (EPA, 2020b). The PA (EPA, 2020b) notes that bias adjustment was not performed in the modeling effort. In their analyses, Lefohn et al. (2014) noted that model performance at low-elevation sites tended toward larger under prediction biases in the cool months (i.e., November-April) and larger over prediction biases in warm months (June-October), particularly for sites in the southern and eastern U.S. For almost all high-elevation sites in their analyses, the model under predicted in the spring months when the above-median MDA8 Global Background O_3 coincided with diagnosed STT-S events as per the stratospheric trajectory analyses performed by Dr. Heini Wernli, Institute for Atmospheric and Climate Science, ETH Zurich in Switzerland. For urban sites with higher spring anthropogenic influences, it was more likely that over predictions were associated with anthropogenic O_3. There continues to be strong evidence, as supported in the literature, as well as EPA's own analyses (e.g., EPA, 2014a), that background O_3 across the U.S. 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.

In summary, empirical data indicate that as emission reductions occur across the U.S., the higher MDA8 concentrations shift at many O_3 monitoring sites from the summer toward the March-June months. In addition, as emission reductions occur, the distribution of hourly average concentrations shift from the higher values toward the middle values and the lower values shift upward toward the middle values. There is a compression of the distribution. Further, as emission reductions occur, background O_3 concentrations increase their percentage in the observed total O_3 concentration with the result that the compressed distribution of hourly average concentrations begins to resemble the distribution of background O_3. Hopefully, it one were to apply a bias adjustment to the estimated modeled USB estimates described in the PA (EPA, 2020b) patterns would result that resemble the background O_3 patterns published previously in the literature, as well as the patterns observed in the empirical data as emission reductions have occurred. If bias adjustments to the model do not change the seasonal patterns described for the current EPA USB model described in the PA (EPA, 2020b), then further model sensitivity analyses should be undertaken. The patterns described earlier that are derived from empirical data (i.e., the compressed distributions and the seasonal shift from the summer months to the March-June period) provide those of us who have applied models to estimate USB, USB_{AB}, or EIB with the opportunity to assess the adequacy of our results. As mentioned earlier, background O_3 plays an important role in the Agency's risk analyses. In turn, the risk analyses play an important role in the Administrator's margin of safety determination for the human health O_3 standard.

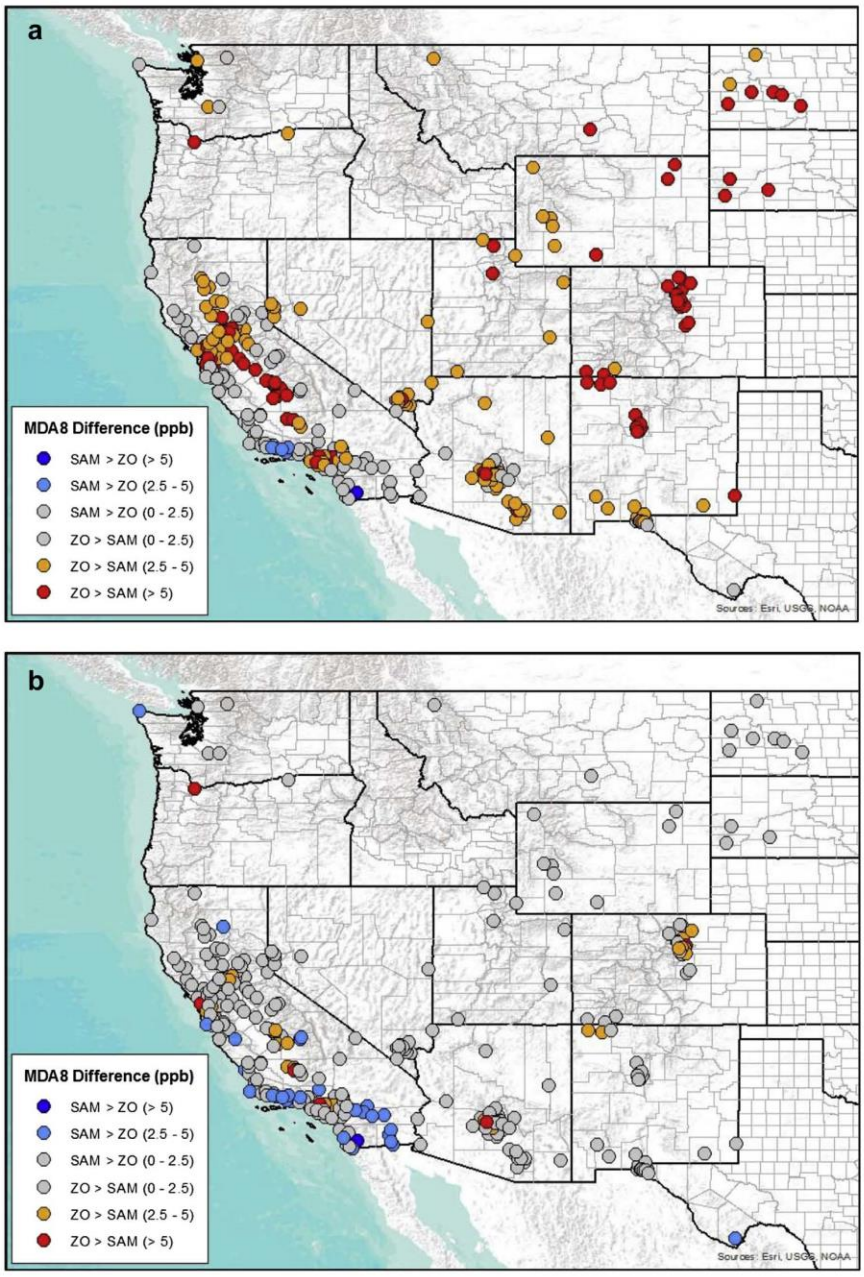


Fig. 5. Difference (ppb) in unadjusted April–October mean MDA8 USB ozone vs mean MDA8 USB_{AB} ozone at monitoring locations across the western U.S. Brighter colors indicate sites where zero out (ZO) estimates of USB exceed source apportionment (SAM) estimates of USB_{AB}. b. Difference (ppb) in bias-adjusted April–October mean MDA8 USB ozone vs. bias-adjusted mean MDA8 USB_{AB} ozone at monitoring locations across the western U.S. Brighter colors indicate sites where zero out (ZO) estimates of USB exceed source apportionment (SAM) estimates of USB_{AB}. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Figure 3-58. Difference between the estimates of USB and USB_{AB}, before and after a bias adjustment is made. Source: Dolwick et al. (2015).

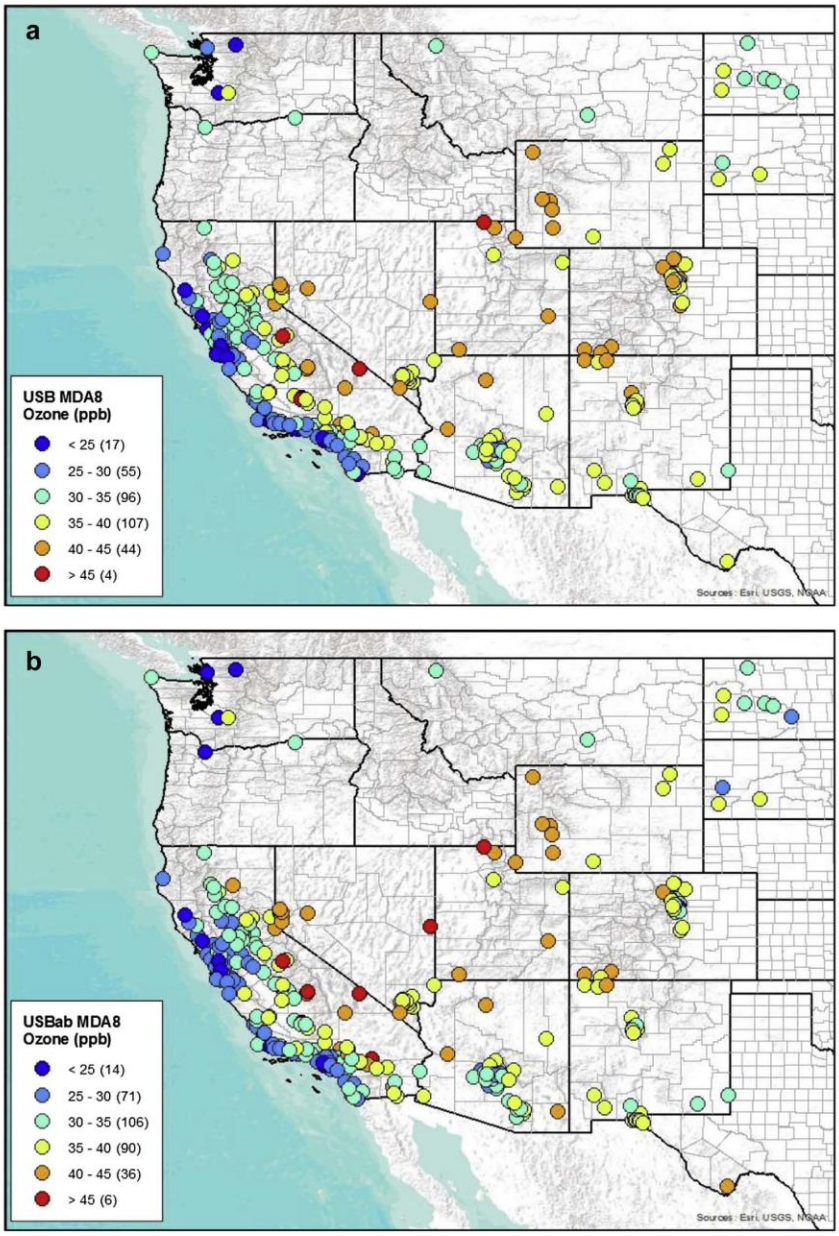


Fig. 6. April–October mean bias-adjusted USB MDA8 ozone (ppb) at monitoring locations across the western U.S., as estimated by a 2007 CMAQ zero out simulation. b. April–October mean bias-adjusted USB_{AB} MDA8 ozone (ppb) at monitoring locations across the western U.S., as estimated by a 2007 CAMx source apportionment simulation.

Figure 3-59. April-October mean bias-adjusted USB MDA8 O_3 (ppb) at monitoring locations across the western U.S., as estimated by a 2007 CMAQ zero-out simulation. b. April-October mean bias-adjusted USB_{AB} MDA8 O_3 (ppb) at monitoring locations across the western U.S., as estimated by a 2007 CAMx source-apportionment simulation. Source: Dolwick et al. (2015).

3.2.11 Background O_3 and the W126 Exposure Metric

The Lapina et al. (2014) analysis is summarized in the PA (EPA, 2020b, pages 2-32, 2-34, 2-35, 2-66) for discussing the contribution for background O_3 to W126 cumulative exposures. Lapina et al. (2014), using three regional or global chemical transport models, quantified the W126 exposure index in the U.S. in the absence of North American anthropogenic emissions (North American background or "NAB"). The investigators noted that the season with the highest observed O_3 concentrations depends on a specific location. According to the authors, this made modeling the maximum 3-month W126 cumulative value in the continental U.S. computationally expensive. To avoid this expense, Lapina et al. (2014) focused on a fixed 3-month period, May–July 2010 for their analyses. However, as noted in previous sections, the maximum 3-month W126 exposures occur during the March-June period in many vegetation areas across the U.S. To better quantify the importance of background O_3 in influencing the W126 cumulative exposures, the estimates performed in the 2014 PA (EPA, 2014a) may provide more insight on the importance of background O_3 influencing the W126 values than the analyses performed by Lapina et al. (2014).

In the 2014 PA (EPA, 2014a), the EPA used the 2007 zero-out modeling to assess NB (i.e., natural background), NAB (North American Background), and USB influences at four sample locations: Atlanta GA, Denver CO, Farmington NM, and Riverside CA. Each of the four analyses locations had relatively high observed values of W126 in 2007, as averaged over all sites in the area: Atlanta (25.1 ppm-hrs), Denver (19.6 ppm-hrs), Farmington (20.2 ppm-hrs), and Riverside (36.0 ppm-hrs). EPA considered the fractional influence of background O_3 on annual W126 levels in four locations. Fig. 3-62 (originally Fig.2-16 in the 2014 PA) shows the results. Based on the fractional influence methodology, natural background sources were estimated to contribute 29-50% of the total modeled W126, with the highest relative influence in the intermountain western U.S. (i.e., Farmington, NM) and the lowest relative influence in the eastern U.S. (i.e., Atlanta). U.S. background (USB) was estimated to contribute 37-65% of the total modeled W126. As noted in the 2014 PA (EPA, 2014a), the proportional impacts of background were slightly less for the W126 metric than for seasonal mean MDA8 (discussed in section 2.4.2 of the 2014 PA), because of the sigmoidal weighting function that places more emphasis on higher O_3 days when background fractions were generally lower. The key conclusion from the EPA's cursory analysis summarized in the 2014 PA (EPA, 2014a) was that background O_3 could comprise a non-negligible portion of current W126 levels across the U.S. These fractional influences were greatest in the intermountain western U.S. and were slightly smaller than the seasonal mean MDA8 metric.

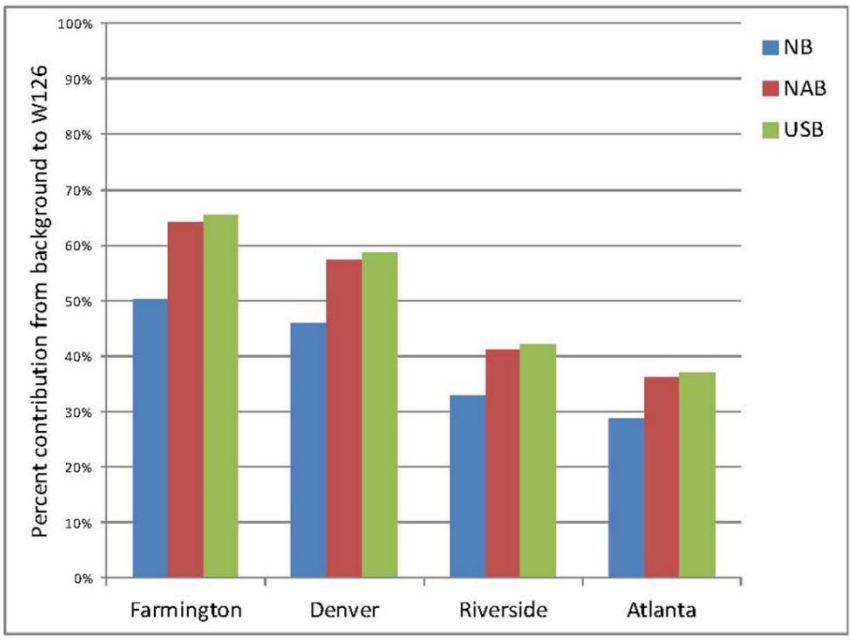


Figure 2-16. Fractional influence of background sources to W126 levels in four sample locations. Model estimates based on 2007 CMAQ zero-out modeling.

Figure 3-62. Fractional influence of background sources to W126 levels in four sample locations. Model estimates based on 2007 CMAQ zero-out modeling. Source: EPA (2014a).

4. Future Research on Health

Areas of future health research are discussed in the PA (Section 3.6) starting on page 3-100. 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, for 6.6 hours while engaged in moderate exertion.**

As noted in the PA (EPA, 2020b), 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 lower MDA8 concentration range. Thus, suggesting

that researchers utilize O_3 concentrations in the background concentration part of the distribution may not provide helpful information for the standard-setting process. I would suggest that additional focus be on the 6.6-hour studies in the 50 ppb range using variable exposure methodologies that resemble current ambient levels.

2. **Epidemiologic studies assessing the influence of "long-term" or "short-term" O_3 exposures is complicated by a lack of knowledge regarding 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 short-term high-level exposure versus a repeated long-term low-level exposure, or a combination of both short-term high-level and repeated long-term low-level exposures, influence health outcomes of the study subjects. Epidemiologic studies that include exposure measurements across a longer-term assessment period and can simultaneously assess the impact of these various elements of exposure (i.e., magnitude, frequency, durations, and pattern) 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 standard 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 reduced the higher concentrations that concentrations of concern for "chronic" effects would also be reduced (Federal Register, 2015 – page 65399). The 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 influence 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, be aware that a long-term epidemiological study does not have to employ exposure metrics based on long-term average concentrations. As shown earlier, EPA noted in the PA (EPA, 2020b) that for sites experiencing emission reductions, exposure metrics that use averaging over longer time periods of hourly O_3 measurements, such as the 6-month (April-September) average of daytime (8am-7pm) O_3 concentrations, show inconsistent trends with only about half of the sites exhibiting decreases in this metric and most other sites exhibiting no trend. Earlier in these comments, it was shown, using the same hourly data, that sites experiencing emission reductions have annual average O_3 concentrations showing increasing trends, while the 4th highest MDA8 values experience decreasing trends. This behavior was associated with the lower hourly average concentrations shifting upward toward the mid-values as emission reductions occurred. Thus, because of this behavior, long-term average exposure metrics appear to have serious limitations for assessing risks associated with O_3 exposures. Both annual average and other long-term average metrics are influenced by the titration of O_3 by NO as NO_x emissions are reduced to protect the public's health and welfare.

Rather, it is suggested that investigators performing long-term epidemiological studies consider using an exposure metric focused on repeated acute exposures whose effects accumulate over time. One example of such a metric was described by Lefohn, Hazucha, Shadwick, and Adams (2010). The authors 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 O_3 concentrations over the mid- and low-range values) on an hourly basis. The weighting scheme focused on the use of daily O_3 exposures that were integrated over time. The authors described a W90 exposure index for use in assessing FEV_1 decrements. The scheme is shown in Fig. 3-63 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 O_3 concentration in units of ppb. The W90 index has units of ppb-hrs.

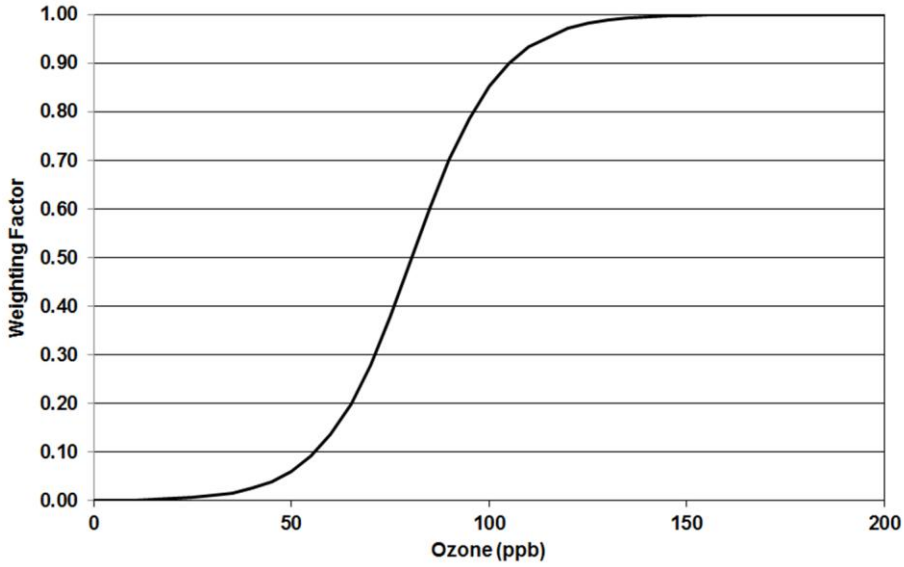


Figure 3-63. The weighting applied to hourly average ozone values for the calculation of the W90 exposure index (see Lefohn et al., 2010).

5. References

Adams, W.C.; Ollison, W.M. 1997. Effects of prolonged simulated ambient ozone dosing patterns on human pulmonary function and symptomatology. Pittsburgh, PA: Air & Waste Management Association.

Adams, W.C. 2003. Comparison of chamber and face mask 6.6-hour exposure to 0.08 ppm ozone via square-wave and triangular profiles on pulmonary responses. Inhalation Toxicology 15: 265-281.

Adams, W.C. 2006a. Comparison of chamber 6.6-h exposures to 0.04 - 0.08 ppm ozone via square-wave and triangular profiles on pulmonary responses. Inhalation Toxicology 18, 127-136.

Adams, W.C. 2006b. Human pulmonary responses with 30-minute time intervals of exercise and rest when exposed for 8 hours to 0.12 ppm ozone via square-wave and acute triangular profiles. Inhalation Toxicology 18, 413-422.

Akriditis, D., Zanis, P., Pytharoulis, I., Mavrakis, A., Karacostas, Th. 2010. A deep stratospheric intrusion event down to the earth's surface of the megacity of Athens. Meteorology and Atmospheric Physics 109, 9-18.

ALA, 2020. American Lung Association. State of the Air 2020. American Lung Association, Washington, DC. https://www.lung.org/our-initiatives/healthy-air/sota/.

Altshuller, A.P, Lefohn, A.S. 1996. Background ozone in the planetary boundary layer over the United States. J Air Waste Manag Assoc 46: 134-141. http://dx.doi.org/10.1080/10473289.1996.10467445.

Amiro, B.D., Gillespie, T.J., Thurtell, G.W. 1984. Injury response of Phaseolus vulgaris to ozone flux density. Atmos. Environ. 18: 1207-1215.

Ashmore, M.R. 1984. Effects of ozone on vegetation in the United Kingdom. Proceedings of the OECD workshop on ozone; February 29-March 2, 1984, Goteburg, Sweden. Goteburg, Sweden: Swedish Environmental Research Institute.

Bennett, J.P., Oshima, R.J., Lippert, L.F. 1979. Effects of ozone on lnJury and dry matter partitioning in pepper plants. Environ. Exp. Bot. 19: 33-39.

Bergweiler, C.J., Manning, W.J. Chevone, B.I. 2008. Seasonal and diurnal gas exchange differences in ozone-sensitive common milkweed (Asclepias syriaca L.) in relation to ozone uptake. Environ Pollut 152: 403-415. http://dx.doi.org/10.1016/j.envpol.2007.06.019.

Blanchard, C.L., Shawb, S.L., Edgerton, E.S., Schwab, J.L. 2019. Emission influences on air pollutant concentrations in New York State: I. ozone. Atmos. Environ. X3 (2019) 10033: 1-12. https://doi.org/10.1016/j.aeaoa.2019.100033.

Blanchard, C.L., Hidy, G.M. 2019. Ozone response to emission reductions in the southeastern United States. Atmos. Chem. Phys., 18, 8183–8202, 2018. https://doi.org/10.5194/acp-18-8183-2018.

Chen, L.-W.A., Chien, L.-C., Li, Y., Lin, G. 2020. Nonuniform impacts of COVID-19 lockdown on air quality over the United States. Science of the Total Environment 745. https://doi.org/10.1016/j.scitotenv.2020.141105.

Cooper, O.R., Stohl, A., Hübler, G., Hsie, E.Y., Parrish, D.D., Tuck, A.F., Kiladis, G.N., Oltmans, S.J., Johnson, B.J., Shapiro, M., Moody, J.L., Lefohn, A.S. 2005. Direct transport of mid-latitude stratospheric ozone into the lower troposphere and marine boundary layer of the tropical Pacific Ocean. Journal of Geophysical Research 110, D23310, doi:10.1029/2005JD005783.

Cox, LA. 2020. Letter from Louis Anthony Cox, Jr., Chair, Clean Air Scientific Advisory Committee, to Administrator Andrew R. Wheeler. Re:CASAC Review of the EPA's Policy Assessment for the Review of the Ozone National Ambient Air Quality Standards (External Review Draft – October 2019). February 19, 2020. EPA-CASAC-20-003. Office of the Adminstrator, Science Advisory Board Washington, DC Available at: https://yosemite.epa.gov/sab/sabproduct.nsf/264cb1227d55e02c85257402007446a4/4713D217BC0710348525851500 6359BA/$File/EPA-CASAC-20-003.pdf.

Cristofanelli, P., Bonasoni, P., Tositti, L., Bonafè, U., Calzolari, F., Evangelisti, F., Sandrini, S., Stohl, A. 2006. A 6-year analysis of stratospheric intrusions and their influence on ozone at Mt. Cimone (2165 m above sea level). Journal of Geophysical Research 111, D03306, doi:10.1029/2005JD006553.

Cristofanelli, P., Bracci, A., Sprenger, M., Marinoni, A., Bonafè, U., Calzolari, F., Duchi, R., Laj, P., Pichon, J.M., Roccato, F., Venzac, H., Vuillermoz, E., Bonasoni, P. 2010. Tropospheric ozone variations at the Nepal Climate Observatory-Pyramid (Himalayas, 5079ma.s.l.) and influence of deep stratospheric intrusion events. Atmospheric Chemistry and Physics 10, 6537–6549. doi:10.5194/acp-10-6537-2010.

Dai, L., Feng, Z., Pan, X., Xu, Y., Li, P., Lefohn, A.S., Harmens, H. Kobayashi, K. 2019. The detoxification by apoplastic antioxidants is insufficient to remove the harmful effects of elevated ozone in tobacco, soybean and poplar. Environ. Pollut. 245: 380-388. DOI: https://doi.org/10.1016/j.envpol.2018.11.030.

Danielsen, E.F. 1968. Stratospheric-tropospheric exchange based on radioactivity, ozone and potential vorticity. J. Atmos. Sci. 25, 502-518.

Danielsen, E.F. 1974; The natural stratosphere of 1974, CIAP Monogr. I, edited by Grobecker A.J., pp 155-122. DOT-TST-75-51. US Dept. of Transp., Washington, D.C.

Danielsen, E.F., Mohnen, V.A. 1977. Project Duststorm report: Ozone transport, in situ measurements and meteorological analyses of tropopause folding. J. Geophys. Res. 82, 5867-5877.

Dantas, G., Siciliano, B., França, B.B., da Silva, C.M., Arbillaa, G. 2020 The impact of COVID-19 partial lockdown on the air quality of the city of Rio de Janeiro, Brazil. Science of the Total Environment 729 (2020). https://doi.org/10.1016/j.scitotenv.2020.139085.

Davies, T.D., Schuepbach, E. 1994. Episodes of high ozone concentrations at the earth's surface resulting from transport down from the upper troposphere/lower stratosphere: a review and case studies. Atmospheric Environment 28, 53-68.

Dolwick, P., Akhtar, F., Baker, K., Possiel, N., Simon, H., Tonnesen, G. 2015. Comparison of background ozone estimates over the western United States based on two separate model methodologies. Atmospheric Environment 109: 282-296, doi: 10.1016/j.atmosenv.2015.01.005.. http://dx.doi.org/10.1016/j.atmosenv.2015.01.005.

Duncan, B. N., Lamsal, L. N., Thompson, A. M., Yoshida, Y., Lu, Z., Streets, D. G., Hurwitz, M. M., Pickering, K. E. 2016. A space-based, high-resolution view of notable changes in urban NOx pollution around the world (2005–2014), J. Geophys. Res. Atmos., 121, 976–996. doi:10.1002/2015JD024121.

Emberson, L., Ashmore, M.R., Cambridge, H.M., Simpson, D., Tuovinen, J.P. 2000a. Modelling stomatal ozone flux across Europe. Environ Pollut 109: 403-413. http://dx.doi.org/10.1016/S0269-7491(00)00043-9.

Emberson, L.D., Wieser, G., Ashmore, M.R. 2000b. Modelling of stomatal conductance and ozone flux of Norway spruce: Comparison with field data. Environ Pollut 109: 393-402. http://dx.doi.org/10.1016/S0269-7491(00)00042-7.

Emery, C., Jung, J., Downey, N., Johnson, J., Jimenez, M., Yarwood, G., Morris, R. 2012. Regional and global modeling estimates of policy relevant background ozone over the United States. Atmospheric Environment 47, 206-217.

Evans, G, Finkelstein, P., Martin, B., Possiel, N., Graves, M. 1983. Ozone measurements from a network of remote sites. Journal of the Air Pollution Control Association, 33, No. 4, 291-296. DOI: 10.1080/00022470.1983.10465575.

Federal Register. Vol. 72, No. 132 / Wednesday, July 11, 2007. National Ambient Air Quality Standards for Ozone, 40 CFR Part 50, 51, 52, 53, and 58, pp 37818-37919.

Federal Register. Vol. 73, No. 60 / Thursday, March 27, 2008. National Ambient Air Quality Standards for Ozone, 40 CFR Part 50, 51, 52, 53, and 58, pp 16436-16514.

Federal Register. Vol. 80, No. 206 / Monday, October 26, 2015. National Ambient Air Quality Standards for Ozone, 40 CFR Part 50, 51, 52, 53, and 58, pp 65292-65468.

Federal Register. Vol. 85, No. 158 / Friday, August 14, 2020. Proposed National Ambient Air Quality Standards for Ozone, 40 CFR Part 50. pp. 49830-49917.

Feng, Z., Büker, P., Pleijel, H., Emberson, L., Karlsson, P.E., Uddling, J. 2017. A unifying explanation for variation in ozone sensitivity among woody plants. Global Change Biol 24: 78-84. http://dx.doi.org/10.1111/gcb.13824.

Fiore, A.M., Oberman, J.T., Lin, M.Y., Zhang, L., Clifton, O.E., Jacob, D.J., Naik, V., Horowitz, L.W., Pinto, J.P., Milly, G.P. 2014. Estimating North American background ozone in U.S. surface air with two independent global models: Variability, uncertainties, and recommendations. Atmos Environ 96: 284-300. http://dx.doi.org/10.1016/j.atmosenv.2014.07.045

FLAG. 2010. Federal Land Managers' Air Quality Related Values Work Group (FLAG). Phase I Report-Revised (2010). Natural Resource Report NPS/NRPC/NRR—2010/232. National Park Service, U.S. Department of the Interior.

Folinsbe,e L.J., McDonnell, W.F., Horstman, D.H. 1988. Pulmonary function and symptom responses after 6.6-hour exposure to 0.12 ppm ozone with moderate exercise. *J Air Waste Manag Assoc* 38: 28-35.

Fowler, D., Cape, J.N. 1982. Air pollutants in agriculture and horticulture. In: Unsworth MH, Ormrod DP. eds. Effects of Gaseous Air Pollution in Agriculture and Horticulture. London: Butterworth Scientific.

Frey, H.C. 2014. Letter from Dr. H. Christopher Frey, Chair, Clean Air Scientific Advisory Committee to Honorable Gina McCarthy, Administrator, US EPA. Re: CASAC Review of the EPA's Second Draft Policy Assessment for the Review of the Ozone National Ambient Air Quality Standards. June 26, 2014. EPA-CASAC-14-004. Office of the Administrator, Science Advisory Board Washington, DC. Available at: https://nepis.epa.gov/Exe/ZyPURL.cgi?Dockey=P100JR6F.txt.

Guo, J.J., Fiore, A.M., Murray, L.T., Jaffe, D.A., Schnell, J.L., Moore, C.T., Milly, G.P. 2018. Average versus high surface ozone levels over the continental USA: Model bias, background influences, and interannual variability. Atmos Chem Phys 18: 12123-12140. http://dx.doi.org/10.5194/acp-18-12123-2018.

Grantz, D.A., Zhang, X.J., Massman, W.J., Den Hartog, G., Neumann, H.H., Pederson, J.R. 1995. Effects of stomatal conductance and surface wetness on ozone deposition in field-grown grape. Atmos Environ 29: 3189-3198. http://dx.doi.org/10.1016/1352-2310(95)00129-M.

Grantz, D.A., Zhang, X.J., Massman, W., Delany, A., Pederson, R. 1997. Ozone deposition to a cotton (Gossypium hirsutum L) field: Stomatal and surface wetness effects during the California Ozone Deposition experiment. Agr Forest Meteorol 85: 19-31. http://dx.doi.org/10.1016/S0168-1923(96)02396-9.

Grantz, D.A., Vu, H.B., Heath, R.L., Burkey, K.O. 2013. Demonstration of a diel trend in sensitivity of Gossypium to ozone: a step toward relating O-3 injury to exposure or flux. J Exp Bot 64: 1703-1713. http://dx.doi.org/10.1093/jxb/ert032.

Goldberg, D.L., Anenberg, S.C., Griffin, D., McLinden, C.A., Lu, Z., Streets, D.G. 2020. Disentangling the Impact of the COVID-19 Lockdowns on Urban NO_2 From Natural Variability. Geophysical Research Letters, 47, e2020GL089269. https://doi.org/10.1029/2020GL089269.

Grewe, V. 2013. A generalized tagging method. Geosci Model Dev 6(1): 247-253.

Grulke, N.E., Alonso, R., Nguyen, T., Cascio, C., Dobrowolski, W. 2004. Stomata open at night in pole-sized and mature ponderosa pine: Implications for O3 exposure metrics. Tree Physiol 24: 1001-1010. http://dx.doi.org/10.1093/treephys/24.9.1001.

Haagenson, P.L., Shapiro, M.A., Middleton, P., Laird, A.R. 1981. A case study relating high ground level ozone to enhanced photochemistry and isentropic transport from the stratosphere. J. Geophys. Res. 86, 5231-5237.

Hazucha, M.J. 1987. Relationship between ozone exposure and pulmonary function changes. Journal of Applied Physiology 62, 1671–1680.

Hazucha, M.J, Folinsbee, L.J., Seal E. 1992. Effects of steady-state and variable ozone concentration profiles on pulmonary function. *Am Rev Respir Dis* **146**: 1487-1493.

Hazucha, M., Lefohn, A.S. 2007. Nonlinearity in Human Health Response to Ozone: Experimental Laboratory Considerations *Atmospheric Environment*. 41: 4559-4570.

Heath, R.L., Lefohn, A.S., Musselman R.C. 2009. Temporal processes that contribute to nonlinearity in vegetation responses to ozone exposure and dose. *Atmos Environ* **43**: 2919-2928.

Heck, W.W., Dunning, J.A., Hindawi, I.J. 1966. Ozone: nonlinear relation of dose and injury in plants. Science 151, 577–578.

Heck, W.W.; Tingey, D.T. 1971. Ozone time-concentration model to predict acute foliar injury. In: Englund, H. M.; Beery, W. T., eds. Proceedings of the second international clean air congress; December 1970; Washington. DC. New York, NY: Academic Press; pp. 249-255.

Henderson, W.R., Reinert, R.A. 1979. Yield response of four fresh market tomato cultivars after acute ozone exposure in the seedling stage. J. Am. Soc. Hortic. Sci. 104: 754-759.

Henderson, R. 2006. Letter from Dr. Rogene Henderson, Chair, Clean Air Scientific Advisory Committee to Honorable Stephen L. Johnson, Administrator, US EPA. Re: CASAC Peer Review of the Agency's 2nd Draft Ozone Staff Paper October 24, 2006. EPA-CASAC-07-001. Office of the Administrator, Science Advisory Board U.S. EPA HQ, Washington DC. Available at: https://nepis.epa.gov/Exe/ZyPURL.cgi?Dockey=P1000WO7.txt.

Hocking, W.K., Carey-Smith, T., Tarasick, D.W., Argall, P.S., Strong, K., Rochon, Y., Zawadzki, I., Taylor, P.A. 2007. Detection of stratospheric ozone intrusions by wind profiler radars. Nature 450, 281–284.

Hogg, A., Uddling, J., Ellsworth, D., Carroll, M.A., Pressley, S., Lamb, B., Vogel, C. 2007. Stomatal and non-stomatal fluxes of ozone to a northern mixed hardwood forest. Tellus B Chem Phys Meteorol 59: 514-525. http://dx.doi.org/10.1111/j.1600-0889.2007.00269.x.

Hogsett, W.E., Tingey, D.T., Holman, S.R. 1985. A programmable exposure control system for determination of the effects of pollutant exposure regimes on plant growth. *Atmos Environ* **19**: 1135–1145.

Horstman, D.H., Folinsbee, L.J., Ives, P.J., Abdul-Salaam, S., McDonnell, W.F. 1990. Ozone concentration and pulmonary response relationships for 6.6-hour exposures with five hours of moderate exercise to 0.08, 0.10, and 0.12 ppm. *Am J Respir Crit Care Med* **142**: 1158-1163.

Huang, X., Ding, A., Gao, J., Zheng, B., Zhou, D., et al. 2020. Enhanced secondary pollution offset reduction of primary emissions during COVID-19 lockdown in China. National Science Review, nwaa137, https://doi.org/10.1093/nsr/nwaa137.

Jaffe, D.A., Cooper, O.R., Fiore, A.M., Henderson, B.H., Tonnesen, G.S., Russell, A.G., Henze, D.K., Langford, A.O., Lin, M., Moore, T. 2018. Scientific assessment of background ozone over the US: Implications for air quality management. 6. http://dx.doi.org/10.1525/elementa.309.

Junge, C. 1962. Global ozone budget and exchange between stratosphere and troposphere. Tellus 14, 363–377.

Kim, C.S., Alexis, N.E., Rappold, A.G., Kehrl, H., Hazucha, M.J., Lay, J.C., Schmitt, M.T., Case, M., Devlin, R.B., Peden, D.B., Diaz-Sanchez, D. 2011. Lung function and inflammatory responses in healthy young adults exposed to 0.06 ppm ozone for 6.6 hours. Am. J. Respir. Crit. Care Med. 183: 1215-1221. http://dx.doi.org/10.1164/rccm.201011-1813OC.

Köllner, B., Krause, GHM. 2003. Effects of two different ozone exposure regimes on chlorophyll and sucrose content of leaves and yield parameters of sugar beet (Beta Vulgaris L.) and rape (Brassica Napus L.). *Water Air Soil Poll.* **144**: 317–332.

Lamarque, J.-F., Hess, P.G. 1994. Cross-tropopause mass exchange and potential vorticity budget in a simulated tropopause folding. Journal of Atmospheric Science 51, 2246-2269.

Langford, A.O., Aikin, K.C., Eubank, C.S., Williams, E.J. 2009. Stratospheric contribution to high surface ozone in Colorado during springtime. Geophysical Research Letters 36, L12801. http://dx.doi.org/10.1029/2009GL038367.

Langford, A.O., Alvarez, R.J, Brioude, J., Fine, R., Gustin, M.S., Lin, M.Y., Marchbanks, R.D., Pierce, R.B., Sandberg, S.P., Senff, C.J., Weickmann, A.M., Williams, E.J. 2017. Entrainment of stratospheric air and Asian pollution by the convective boundary layer in the southwestern US. J Geophys Res Atmos 122: 1312-1337. http://dx.doi.org/10.1002/2016JD025987.

Lapina, K., Henze, D.K., Milford, J.B., Huang, M., Lin, M., Fiore, A.M., Carmichael, G., Pfister, G.G.,Bowman, K. 2014. Assessment of source contributions to seasonal vegetative exposure to ozone in the US. J. Geophys. Res. Atmos. 119(1): 324-340.

Larsen, R.I., Heagle, A.S., Heck, W.W. 1983. An air quality data analysis system for interrelating effects, standards, and needed source reductions: Part 7. An 03 -502 leaf injury mathematical model. J. Air Pollut. Control Assoc. 33: 198-207.

Larsen, R. I., Heck, W.W. 1984. An air quality data analysis system for interrelating effects, standards, and needed source reductions: Part 8. An effective mean 03 crop reduction mathematical model. J. Air Pollut. Control Assoc. 34: 1023-1034.

Le, T., Wang, Y., Liu, L., Yang, J., Yung, Y.L., Li, G., Seinfeld, J.H. 2020. Unexpected air pollution with marked emission reductions during the COVID-19 outbreak in China. Science 10.1126/science.abb7431.

Lee, E.H., Hogsett, W.E. 1999. Role of concentrations and time of day in developing ozone exposure indices for a secondary standard. *J Air Waste Manage Assoc* **49**: 669–681.

Lefohn, A.S., Benedict, H.M. 1982. Development of a mathematical index that describes ozone concentration, frequency, and duration. Atmospheric Environment 16:2529-2532.

Lefohn, A.S. and Foley, J.K. 1993. Establishing Ozone Standards to Protect Human Health and Vegetation: Exposure/Dose-Response Considerations. J. Air Waste Manag. Assoc. 43(2):106-112.

Lefohn, A.S., Runeckles, V.C. 1987. Establishing standards to protect vegetation - Ozone exposure/dose considerations. Atmospheric Environment 21:561-568.

Lefohn, A.S., Lawrence, J.A., Kohut, R.J. 1988. A comparison of indices that describe the relationship between exposure to ozone and reduction in the yield of agricultural crops. Atmospheric Environment 22:1229-1240.

Lefohn, A.S., Shadwick, D.S., Ziman, S.D. 1998. The difficult challenge of attaining EPA's new ozone standard. Environmental Science & Technology 32(11):276A-282A.

Lefohn, A.S., Oltmans, S.J., Dann, T., Singh, H.B. 2001. Present-day variability of background ozone in the lower troposphere. J. Geophys. Res. 106(D9):9945-9958, doi:10.1029/2000JD900793.

Lefohn, A.S., Hazucha, M.J., Shadwick, D., Adams, W.C. 2010. An alternative form and level of the human health ozone standard. Inhalation Toxicology 22:999-1011.

Lefohn, A.S., Wernli, H., Shadwick, D., Limbach, S., Oltmans, S.J., Shapiro, M. 2011. The importance of stratospheric-tropospheric transport in affecting surface ozone concentrations in the Western and Northern Tier of the United States. Atmospheric Environment 45:4845-4857.

Lefohn, A.S., Wernli, H., Shadwick, D., Oltmans, S.J., Shapiro, M. 2012. Quantifying the frequency of stratospheric-tropospheric transport affecting enhanced surface ozone concentrations at high- and low-elevation monitoring sites in the United States. Atmospheric Environment 62: 646-656.

Lefohn, A.S., Emery, C., Shadwick, D., Wernli, H., Jung, J., Oltmans, S.J. 2014. Estimates of background surface ozone concentrations in the United States based on model-derived source apportionment. Atmospheric Environment 84:275-288. http://dx.doi.org/10.1016/j.atmosenv.2013.11.033.

Lefohn, A. S., Malley, C. S., Simon, H., Wells. B., Xu, X., Zhang, L., Wang, T. 2017. Responses of human health and vegetation exposure metrics to changes in ozone concentration distributions in the European Union, United States, and China. Atmospheric Environment, 152: 123-145. doi:10.1016/j.atmosenv.2016.12.025.

Lefohn, A.S., Malley, C.S., Smith, L., Wells, B., Hazucha, M., Simon, H., Naik, V., Mills, G., Schultz, M.G., Paoletti, E., De Marco, A., Xu, X., Zhang, L., Wang, T., Neufeld, H.S., Musselman, R.C., Tarasick, T., Brauer, M., Feng, Z., Tang, T., Kobayashi, K., Sicard, P., Solberg, S., and Gerosa. G. 2018. Tropospheric ozone assessment report: global ozone metrics for climate change, human health, and crop/ecosystem research. Elem Sci Anth, 6(1):28. DOI: https://doi.org/10.1525/elementa.279.

Lefohn, A.S. 2019a. Comments on the First Draft of the EPA Integrated Scientific Assessment Document (External Review Draft, EPA/600/R-19/093, September 2019) Docket ID No. EPA-HQ-ORD-2018-0274-0034.

Lefohn, A.S. 2019b. Comments on the First Draft of the EPA Policy Assessment for the Review of the Ozone National Ambient Air Quality Standards (External Review Draft, EPA-452/P-19-002, October 2019) Docket ID No. EPA-HQ-OAR-2018-0279-0027.

Lin, M., Fiore, A.M., Cooper, O.R., Horowitz, L.W., Langford, A.O., Levy II, H., Johnson, B.J., Naik, V., Oltmans, S.J., Senff, C.J. 2012a. Springtime high surface ozone events over the western United States: Quantifying the role of stratospheric intrusions. Journal of Geophysical Research 117, D00V22, doi:10.1029/2012JD018151.

Lin, M., Fiore, A.M., Cooper, O.R., Horowitz, L.W., Langford, A.O., et al. 2012. Springtime high surface ozone events over the western United States: Quantifying the role of stratospheric intrusions. *J Geophy Res* **117**: D00V22, doi:10.1029/2012JD018151.

Lu, X., Hong, J., Zhang, L., Cooper, O.R., Schultz, M.G., Xu, X., Wang, T., Gao, M., Zhao, Y., Zhang, Y. 2018. Severe surface ozone pollution in China: a global perspective. Environmental Science & Technology Letters 5 (8), 487-494. DOI: 10.1021/acs.estlett.8b00366.

Ludwig, F.L., Reiter, E., Shelar, E., Johnson, W.B. 1977. The relation of oxidant levels to precursor emissions and meteorological features: v. I, analysis and findings. Research Triangle Park, NC: U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards; report no. EPA-450/3-77-022a. Available from: NTIS, Springfield, VA; PB-275 001.

Mahato, S., Pal, S., Ghosh, K. G. 2020. Effect of lockdown amid COVID-19 pandemic on air quality of the megacity Delhi, India. Science of the Total Environment 730. https://doi.org/10.1016/j.scitotenv.2020.139086.

Male, L., Preston, E., Neely, G. 1983. Yield response curves of crops exposed to $S0_2$ time series. Atmos. Environ. 17: 1589-1593.

Matyssek, R., Sandermann, H., Wieser, G., Booker, F., Cieslik, S., Musselman, R., Ernst, D. 2008. The challenge of making ozone risk assessment for forest trees more mechanistic [Review]. Environ Pollut 156: 567-582. http://dx.doi.org/10.1016/j.envpol.2008.04.017.

McDonald-Buller, E.C., Allen, D.T., Brown, N., Jacob, D.J., Jaffe, D., Kolb, C.E., Lefohn, A.S., Oltmans, S., Parrish, D.D., Yarwood, G., Zhang, L. 2011. Establishing Policy Relevant Background (PRB) Ozone Concentrations in the United States. Environmental Science & Technology. 45(22):9484-97.

McGrath, J.M., Betzelberger, A.M., Wang, S., Shook, E., Zhu, X.G., Long, S.P., Ainsworth, E.A. 2015. An analysis of ozone damage to historical maize and soybean yields in the United States. Proc Natl Acad Sci USA 112: 14390-14395. http://dx.doi.org/10.1073/pnas.1509777112.

McLaughlin, S.B., Shriner, D.S., McConathy, R.K., Mann, L.K. 1979. The effects of $S0_2$ dosage kinetics and exposure frequency on photosynthesis and transpiration of kidney beans (Phaseolus vulgaris L.). Environ. Exp. Bot. 19: 179-191.

Miller, F.J., Schlosser, P.M., Janszen, D.B. 2000. Haber's rule: a special case in a family of curves relating concentration and duration of exposure to a fixed level of response for a given endpoint. *Toxicology* **149** (1): 21-34.

Mills, G., Pleijel, H., Braun, S.,Büker, P., Bermejo, V., et al. 2011. New stomatal flux-based critical levels for ozone effects on vegetation. Atmos Environ 45: 5064-5068. http://dx.doi.org/10.1016/j.atmosenv.2011.06.009.

Mills, G., et al. 2018. Tropospheric Ozone Assessment Report: Present-day tropospheric ozone distribution and trends relevant to vegetation. Elem Sci Anth, 6: 47. DOI: https://doi.org/10.1525/elementa.302.

Musselman, R.C., Oshima, R.J., Gallavan, R.E. 1983. Significance of pollutant concentration distribution in the response of 'red kidney' beans to ozone. *J Am Soc Hortic Sci* **108**: 347–351.

Musselman, R.C., Huerta, A.J., McCool, P.M., Oshima, R.J. 1986. Response of beans to simulated ambient and uniform ozone distributions with equal peak concentration. J Am Soc Hortic Sci 111: 470–473.

Musselman, R.C., Younglove, T., McCool, P.M. 1994. Response of Phaseolus vulgaris L. to differing ozone regimes having identical total exposure. Atmos Environ 28: 2727–2731

Musselman, R.C., Massman, W.J. 1999. Ozone flux to vegetation and its relationship to plant response and ambient air quality standards. Atmos Environ 33: 65-73.

Musselman, R.C., Lefohn, A.S., Massman ,W.J., Heath, R.L. 2006. A critical review and analysis of the use of exposure- and flux-based ozone indices for predicting vegetation effects. *Atmos Environ* 40(10): 1869-1888.

Naranjo, L., "Volatile Trees," NASA.gov, 20 November 2011. Retrieved: 9 November 2019. https://earthdata.nasa.gov/learn/sensing-our-planet/volatile-trees.

Neufeld, H.S., Sullins, A., Sive, B.C., Lefohn, A.S. 2019. Spatial and temporal patterns of ozone at Great Smoky Mountains National Park and implications for plant responses. Atmos. Environ.: X 2, 100023. DOI: https://doi.org/10.1016/j.aeaoa.2019.100023.

Nussbaum, S., Geissmann, M., Fuhrer, J. 1995. Ozone exposure–response relationships for mixtures of perennial ryegrass and white clover depend on ozone exposure patterns. *Atmos Environ* **29**: 989–995.

Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X., Hayasaka, T. 2007. An Asian emission inventory of anthropogenic emission sources for the period 1980–2020, Atmos. Chem. Phys., 7, 4419–4444, doi:10.5194/acp-7-4419-2007.

Oltmans, S.J., Lefohn, A.S., Harris, J.M., Tarasick, DW., Thompson, AM., Wernli, H., Johnson, B.J., Novelli, P.C., Montzka, S.A., Ray, J.D., Patrick, L.C., Sweeney, C., Jefferson, A., Dann, T., Davies, J., Shapiro, M., Holben, B.N. 2010. Enhanced ozone over western North America from biomass burning in Eurasia during April 2008 as seen in surface and profile observations. Atmospheric Environment. 44: 4497-4509.

Ordóñez, C., Brunner, D., Staehelin, J., Hadjinicolaou, P., Pyle, J.A., Jonas, M., Wernli, H., Prevot, A.S.H. 2007. Strong influence of lowermost stratospheric ozone on lower tropospheric background ozone changes over Europe. Geophysical Research Letters 34, L07805, doi:10.1029/2006GL029113.

Oshima, R.J., Braegelmann, P.K., Baldwin, D. W., Van Way, V., Taylor, O.C. 1977a. Reduction of tomato fruit size and yield by ozone. J. Am. Soc. Hortic. Sci. 102: 289-293.

Oshima, R J., Braegelmann, P.K., Baldwin, D.W., Van Way, V., Taylor, O.C. 1977b. Responses of five cultivars of fresh market tomato to ozone: a contrast of cultivar screening with foliar injury and yield. J. Am. Soc. Hortic. Sci. 102: 286-289.

Oksanen, E., Holopaninen, T. 2001. Responses of two birch (Betula pendula Roth. clones to different ozone profiles with similar AOT40 exposure. Atmos Environ 35: 5245–5254.

Paoletti, E., Manning, W.J. 2007. Toward a biologically significant and usable standard for ozone that will also protect plants [Review]. Environ Pollut 150: 85-95. http://dx.doi.org/10.1016/j.envpol.2007.06.037.

Patel, H., Talbot, N., Salmond, J., Dirks, K., Xie, S., Davy, P. 2020. Implications for air quality management of changes in air quality during lockdown in Auckland (New Zealand) in response to the 2020 SARS-CoV-2 epidemic. Science of the Total Env. https://doi.org/10.1016/j.scitotenv.2020.141129.

Reed, R.J. 1955. A study of a characteristic type of upper level frontogenesis. J. Meteorol. 12, 226-237.

Reinert, R.A., Nelson, P.V. 1979. Sensitivity and growth of twelve elatior begonia cultivars to ozone. HortScience 14: 747-748.

Rice, J. 2014. Ozone Monitoring Season Analysis. Memorandum to the Ozone NAAQS Review Docket, EPA–HQ–OAR–2008–0699.

Samet, JM. 2010. Letter from Jonathan Samet, Chair, Clean Air Scientific Advisory Committee, to Administrator Lisa Jackson. Re: CASAC Review of EPA's Proposed Ozone National Ambient Air Quality Standard (Federal Register, Vol. 75, Nov. 11, January 19, 2010). February 19, 2010. EPA-CASAC-10-007. Office of the Administrator, Science Advisory Board U.S. EPA HQ, Washington DC. Available at: **https://nepis.epa.gov/Exe/ZyPURL.cgi?Dockey=P10072T1.txt**.

Schelegle, E.S., Walby, W.F., Adams, W.C. 2007. Time course of ozone-induced changes in breathing pattern in healthy exercising humans. J Appl Physiol. 102:688–697.

Schelegle, E.S., Morales, C.A., Walby, W.F., Marion, S., Allen, R.P. 2009. 6.6-hour inhalation of ozone concentrations from 60 to 87 ppb in healthy humans. Am. J. Respir. Crit. Care Med. 180:265–272.

Schuepbach, E., Davies. T.D., Massacand, A.C. 1999. An unusual springtime ozone episode at high elevation in the Swiss Alps: Contributions both from cross-tropopause exchange and from the boundary layer. Atmospheric Environment 33, 1735-1744.

Schultz, M.G. et al. 2017. Tropospheric Ozone Assessment Report: Database and Metrics Data of Global Surface Ozone Observations. Elementa 5(58). DOI: https://doi.org/10.1525/elementa.244

Shapiro, M.A. 1980. Turbulent mixing within tropopause folds as a mechanism for the exchange of chemical constituents between the stratosphere and troposphere. J. Atmos. Sci. 37, 994-1004.

Sicard, P., Serra, R., Rossello, P. 2016. Spatiotemporal trends in ground-level ozone concentrations and metrics in France over the time period 1999–2012, Environ. Res., 149, 122-144. http://dx.doi.org/10.1016/j.envres.2016.05.014.

Sicard, P., De Marco, A., Agathokleous, E., et al. 2020. Amplified ozone pollution in cities during the COVID-19 lockdown, *Science of the Total Environment*, https://doi.org/10.1016/j.scitotenv.2020.139542.

Simon H, Reff A, Wells B, Xing J, Frank N. 2015. Ozone trends across the United States over a period of decreasing NOx and VOC emissions. *Environ Sci Technol* **49**: 186-195. dx.doi.org/10.1021/es504514z.

Skelly, J.M. 2000. Tropospheric ozone and its importance to forests and natural plant communities of the northeastern United States. Northeastern Naturalist. 7, 221-236.

Škerlak, B., Sprenger, M., Wernli, H. 2014. A global climatology of stratosphere–troposphere exchange using the ERA-Interim data set from 1979 to 2011. Atmospheric Chemistry and Physics, 14, 913–937, doi:10.5194/acp-14-913-2014.

Škerlak, B., Sprenger, M., Pfahl, S., Tyrlis, E., Wernli, H. 2015. Tropopause folds in ERA-Interim: Global climatology and relation to extreme weather events. J Geophys Res Atmos 120: 4860-4877. http://dx.doi.org/10.1002/2014JD022787.

Škerlak, B., Pfhal, S., Sprenger, M., Wernli, H. 2019. A numerical process study on the rapid transport of stratospheric air down to the surface over western North America and the Tibetan Plateau. Atmospheric Chemistry and Physics, 19, 6535–6549, https://doi.org/10.5194/acp-19-6535-2019.

Sommer, L., Hersher, R., Huo, J., Benincasa, R. 2020. Traffic is way down because of lockdown, but air pollution? Not so much. National Public Radio. https://www.npr.org/sections/health-shots/2020/05/19/854760999/traffic-is-way-down-due-to-lockdowns-but-air-pollution-not-so-much.

Stohl, A., Spichtinger-Rakowsky, N., Bonasoni, P., Feldmann, H., Memmesheimer, M., Scheel, H.E., Trickl, T., Hübener, S. 2000. The influence of stratospheric intrusions on alpine ozone concentrations. Atmospheric Environment 34, 1323-1354.

Tonneijck, A.E.G. 1984. Effects of peroxyacetyl nitrate (PAN) and ozone on some plant species. In: Proceedings of the OECD workshop on ozone; Goteborg, Sweden. Goteborg, Sweden: Swedish Environ. Research Institute.

Trainer, M., Parrish, D.D., Buhr, M.P., Norton, R.B., Fehsenfeld, F.C., et al. 1993. Correlation of ozone with NOy in photochemically aged air. J Geophys Res 98: 2917-2925.

Turnipseed, A.A., Burns, S.P., Moore, D.J.P., Hu, J., Guenther, A.B.,Monson, R.K. 2009. Controls over ozone deposition to a high elevation subalpine forest. Agr Forest Meteorol 149: 1447-1459. http://dx.doi.org/10.1016/j.agrformet.2009.04.001.

Uddling, J., Teclaw, R.M., Pregitzer, K.S.,Ellsworth, D.S. 2009. Leaf and canopy conductance in aspen and aspen-birch forests under free-air enrichment of carbon dioxide and ozone. Tree Physiol 29: 1367-1380. http://dx.doi.org/10.1093/treephys/tpp070.

U.S. Court of Appeals FOR THE DISTRICT OF COLUMBIA CIRCUIT Argued December 18, 2018 Decided August 23, 2019 No. 15-1385.

U.S. EPA. 1978. Air quality criteria for ozone and other photochemical oxidants [EPA Report]. (EPA/600/8-78/004). Washington, DC. https://nepis.epa.gov/exe/ZyPURL.cgi?Dockey=200089CW.txt.

U.S. EPA. 1986. Air quality criteria for ozone and other photochemical oxidants [EPA Report]. (EPA-600/8-84-020aF - EPA-600/8-84-020eF). Research Triangle Park, NC. https://ntrl.ntis.gov/NTRL/dashboard/searchResults.xhtml?searchQuery=PB87142949.

U.S. EPA. 1992. Summary of Selected New Information on Effects of Ozone on Health and Vegetation: Supplement to 1986 Air Quality Criteria for Ozone and Other Photochemical Oxidants. Report No. EPA/600/8-88/105F. Research Triangle Park, NC: Environmental Protection Agency. Available from: NTIS, Springfield, VA, PB92-235670.

U.S. EPA. 1996a. Air quality criteria for ozone and related photochemical oxidants [EPA Report]. (EPA/600/P-93/004AF). Research Triangle Park, NC.

U.S. EPA. 1996b. Review of the national ambient air quality standards for ozone: Policy assessment of scientific and technical information: OAQPS staff paper [EPA Report]. (EPA/452/R-96/007). Research Triangle Park, NC. https://www.epa.gov/sites/production/files/2020-07/documents/1996_o3sp_final2.pdf.

U.S. EPA. 2006. Air Quality Criteria for Ozone and Related Photochemical Oxidants (2006 Final). EPA/600/R-05/004aF-cF. Washington, DC: Environmental Protection Agency. Available at: https://cfpub.epa.gov/ncea/risk/recordisplay.cfm?deid=149923.

U.S. EPA. 2007. Review of the national ambient air quality standards for ozone: Policy assessment of scientific and technical information: OAQPS staff paper [EPA Report]. (EPA/452/R-07/007). Research Triangle Park, NC. https://www3.epa.gov/ttn/naaqs/standards/ozone/data/2007_07_ozone_staff_paper.pdf.

US EPA. 2013. Integrated Science Assessment of Ozone and Related Photochemical Oxidants (Final Report). EPA/600/R-10/076F. Research Triangle Park, NC: Environmental Protection Agency. Available at: http://www.epa.gov/ttn/naaqs/standards/ozone/s_o3_2008_isa.html.

U.S. EPA. 2014a. Policy Assessment for the Review of the Ozone National Ambient Air Quality Standards. Final Report. EPA-452/R-14-006. Research Triangle Park, NC: Office of Air Quality Planning and Standards. August.

US EPA. 2014b. Health Risk and Exposure Assessment for Ozone. Final Report. EPA/452/R-14-004a. Research Triangle Park, NC: Environmental Protection Agency. Available at: https://www.epa.gov/naaqs/ozone-o3-standards-risk-and-exposure-assessments-current-review.

US. EPA. 2014c. Welfare Risk and Exposure Assessment for Ozone. Final Report. EPA-452/P-14-005a. Research Triangle Park, NC: Environmental Protection Agency. Available at https://www.epa.gov/naaqs/ozone-o3-standards-risk-and-exposure-assessments-current-review.

US. EPA. 2014d. Health Risk and Exposure Assessment for Ozone Final Report Chapters 7-9 Appendices. EPA-452/R-14-004e. Research Triangle Park, NC: Environmental Protection Agency. Available at https://www.epa.gov/naaqs/ozone-o3-standards-risk-and-exposure-assessments-review-completed-2015.

U.S. EPA. 2015. Implementation of the 2015 Primary Ozone NAAQS: Issues associated with background ozone. White paper for discussion. Research Triangle Park, NC: U.S. Environmental Protection Agency: U.S. EPA.

U.S. EPA. 2018. Technical Assistance Document for the Reporting of Daily Air Quality – the Air Quality Index (AQI). EPA 454/B-18-007. Research Triangle Park, NC: Environmental Protection Agency. Available at https://www3.epa.gov/airnow/aqi-technical-assistance-document-sept2018.pdf.

US EPA. 2019a. Integrated Science Assessment (ISA) for Ozone and Related Photochemical Oxidants (External Review Draft). EPA/600/R-19/093. September. Available at https://cfpub.epa.gov/ncea/isa/recordisplay.cfm?deid=344670.

US EPA. 2019b. Policy Assessment for the Review of the Ozone National Ambient Air Quality Standards, External Review Draft. EPA-452/P-19-002. October. Available at https://www.epa.gov/sites/production/files/2019-10/documents/o3-draft_pa-oct31-2019-erd.pdf.

U.S. EPA, 2019c: A Look Back: Ozone in 2018. https://epa.maps.arcgis.com/apps/Cascade/index.html?appid=9bec4031ba6f4887a9f332a8f058b198

US EPA. 2020a. Integrated Science Assessment of Ozone and Related Photochemical Oxidants. EPA/600/R-20/012. April. Research Triangle Park, NC: Environmental Protection Agency. April. Available at: https://www.epa.gov/naaqs/ozone-o3-standards-integrated-science-assessments-current-review.

US EPA. 2020b. Policy Assessment for the Review of the Ozone National Ambient Air Quality Standards. EPA-452/R-20-001. May. Available at https://www.epa.gov/naaqs/ozone-o3-standards-policy-assessments-current-review.

U.S. EPA, 2020c. April 1, 2020 letter from EPA Administrator Andrew R. Wheeler to Dr. Louis Anthony Cox, Jr., Chair of CASAC.

Wang, L., Pang, J., Feng, Z., Zhu, J., Kazuhiko, K. 2015. Diurnal variation of apoplastic ascorbate in winter wheat leaves in relation to ozone detoxification. *Environ Pollut* **207**: 413-419.

Wang, M., Aaron, C.P., Madrigano, J., et al. 2019. Association between long-term exposure to ambient air pollution and change in quantitatively assessed emphysema and lung function. JAMA. 2019;322(6):546-556. doi:10.1001/jama.2019.10255.

Wang, X., Zheng, O., Feng, Z., Xie, J., Feng, Z., Ouyang, Z., Manning, W.J. 2008. Comparison of a diurnal vs steady-state ozone exposure profile on growth and yield of oilseed rape (Brassica napus L.) in open-top chambers in the Yangtze Delta, China. Environmental Pollution 156 (2008) 449-453.

Wernli, H., Davies, H.C. 1997. A Lagrangian-based analysis of extratropical cyclones. I: The method and some applications. Quart. J. Roy. Meteor. Soc. 123, 467-489.

Wernli, H., Bourqui, M. 2002. A Lagrangian "one-year climatology" of (deep) cross-tropopause exchange in the extratropical northern hemisphere. J. Geophys. Res. 107(D2), 4021, doi:10.1029/2001JD000812.

Xu, X., et al. 2020. Long-term changes of regional ozone in China: implications for human health and ecosystem impacts. Elem Sci Anth, 8: 13. DOI: https://doi.org/10.1525/elementa.409.

Xue, L., Wang, T., Louie, P.K.K., Luk, C.W.Y., Blake, D.R., Xu, Z. 2014. Increasing External Effects Negate Local Efforts to Control Ozone Air Pollution: A Case Study of Hong Kong and Implications for Other Chinese Cities, Environ. Sci. Technol., 48, 10769−10775, dx.doi.org/10.1021/es503278g.

Yun, S-C, Laurence, J.A. 1999. The response of sensitive and tolerant clones of Populus tremuloides to dynamic ozone exposure under controlled environmental conditions. New Phytologist 143, 305–313.

Zhang, L, Jacob, D.J., Downey, N.V., Wood, D.A., Blewitt, D., Carouge, C.C., van Donkelaar, A., Jones, D.B.A., Murray, L.T., Wang, Y. 2011. Improved estimate of the policy-relevant background ozone in the United States using the GEOS-Chem global model with 1/2° × 2/3° horizontal resolution over North America. Atmospheric Environment 45, 6769-6776.