

Background Ozone and Its Importance in Relation to the Health Risk and Exposure Assessment for Ozone Assessment Document

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Executive Summary

Background O₃ is of interest for the following two reasons: (1) at times background O₃ is associated with high concentrations (e.g., exceptional events in the Intermountain West) and (2) background O₃ contributes on a continuous basis to the distribution of observed hourly average O₃ concentrations and affects risk estimates and the attainability of the O₃ NAAQS. In our previous comments to EPA (Lefohn and Oltmans, 2012), we emphasized the importance of background O₃ in relation to health risk and exposure assessments and the role it plays in affecting risk and exposure estimates.

In its second draft of the REA, the Agency has made the decision to use total risk in its estimates and believes that background O₃ is fully represented in these estimates given that the measured and adjusted air quality concentrations being used in the risk and exposure analyses include O₃ produced from precursor emissions from both anthropogenic and background sources. Although the EPA has not discussed the relative importance of background O₃ in the REA, we believe that the Agency has provided information in both the REA and PA that allows one to assess the relative contribution of (1) anthropogenic (i.e., controllable) O₃ and (2) background O₃ (non controllable) to its human health risk estimates.

As mentioned above by the EPA, the distribution of total O₃ concentrations consist of both anthropogenic and background contributions. Background O₃ distributions contribute to the risk estimates more and more as emissions are reduced to attain the various levels of existing and alternative O₃ standards. This is because as more stringent O₃ levels are attained, O₃ concentrations associated with anthropogenic emissions contribute less of a percentage to total O₃ than background O₃ concentrations. Simulation of just meeting the existing and alternative O₃ standards is accomplished by adjusting hourly O₃ concentrations measured over the O₃ season using a model-based adjustment methodology that estimates O₃ sensitivities to precursor emissions changes. These sensitivities, which estimate the response of O₃ concentrations to reductions in anthropogenic NO_x and VOC emissions, are developed using the Higher-order Decoupled Direct Method (HDDM) capabilities in the Community Multi-scale Air Quality (CMAQ) model.

Background O₃ concentrations vary by the hour and are generally highest during the springtime at many O₃ monitoring sites across the US (see Lefohn et al. [2014a] for a discussion of emission-influenced background or referred to as apportionment-based US background in the PA). As indicated in Fig. E1, a large percentage (i.e., >50%) of the 8-hour concentrations measured across the US consists of background O₃. In the West, Intermountain West, and the Northeast, the percentage contribution of background O₃ to the seasonal mean 8-hour concentration is 70% or greater. The information provided by EPA in the PA in Fig. E1 agrees in general with the percent contributions of background O₃ presented by Lefohn et al. (2014a) in their recent publication in *Atmospheric Environment*, which provided the characterization of background O₃ for 23 urban- and rural-influenced sites across the US. Summary figures from Lefohn et al. (2014a) are presented in our comments.

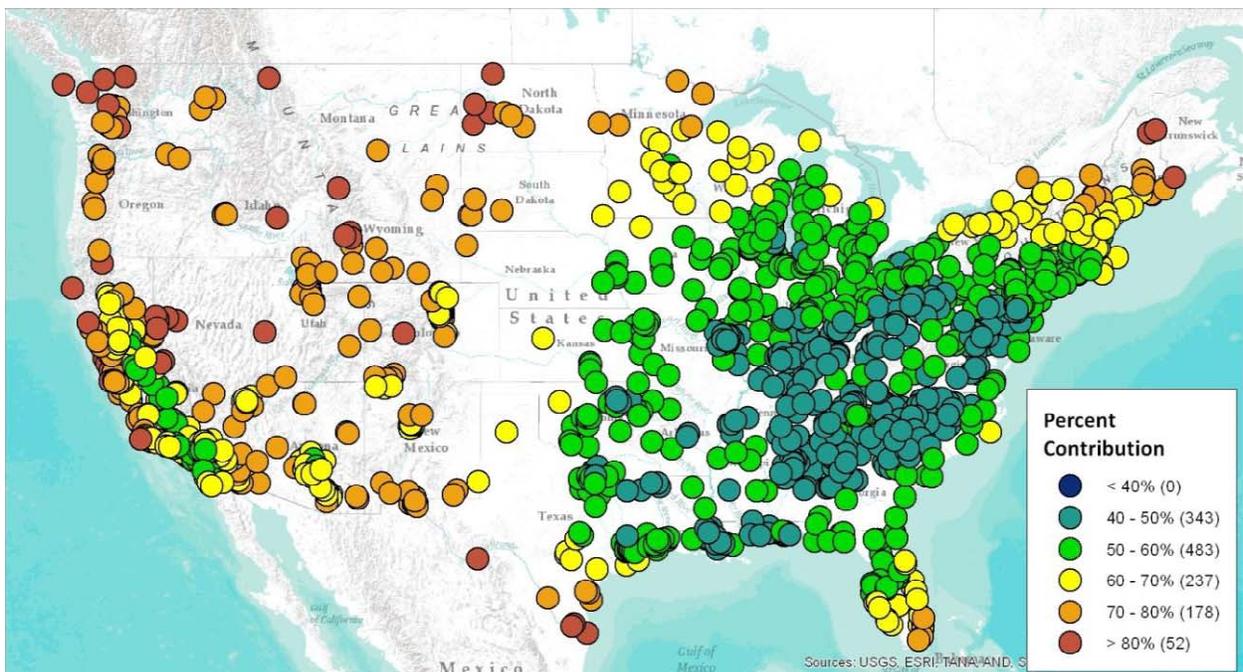


Fig. E1. Map of apportionment-based U.S. background percent contribution to seasonal mean O₃ based on 2007 CAMx source apportionment modeling. (Source: page 2-18 of PA).

As observed in Fig. E1 above (from EPA's PA), background O₃ makes up a large percentage of the monitored observed O₃ concentrations and therefore, plays an important role in comparing the REA risk outcomes across base conditions, attaining the current standard, and attaining alternative standard levels.

One of the important key elements associated with the risk estimates is the change in the distribution of O₃ concentrations between recent O₃ concentrations and adjusted (meeting the existing or alternative standards O₃ scenarios). With respect to the changes in the distribution of O₃ concentrations as a function of emission reductions, the risk metrics used in the REA were influenced by how the distribution of O₃ concentrations change (REA, page 9-32). The change in the distribution of hourly average O₃ concentrations results from emission reductions. These

emissions reductions change the distribution of O₃ concentrations in the high-, mid-, and low-range values.

How distributions of O₃ concentrations change as emissions are reduced has been documented in the literature. Lefohn et al. (1998) noted that as O₃ levels improved (i.e., the environment experienced lower O₃ exposures) due to reduced emissions, reductions in the number of high hourly average concentrations, as well in the number of low hourly average concentrations occurred. The reduction in the number of low hourly average O₃ concentrations was associated with lack of NO_x scavenging (US EPA, 1996). Lefohn et al. (1998) predicted that as a site's air quality improved, the distribution of the hourly average concentrations would move from both the high end as well as the low end of the distribution toward the center (i.e., 30-60 ppb). Several investigators (Oltmans et al., 2006; Lefohn et al., 2008; Oltmans et al., 2008; Lefohn et al., 2010; Oltmans et al., 2013) have reported trending results that showed shifts in the distribution over time, where both the high and the low ends of the distributions of O₃ concentrations shifted toward the mid-level values. Fig. E2 from Lefohn et al. (2010) illustrates the concentrations shifts by month for a site in San Bernardino (CA) for the period 1994-2008.

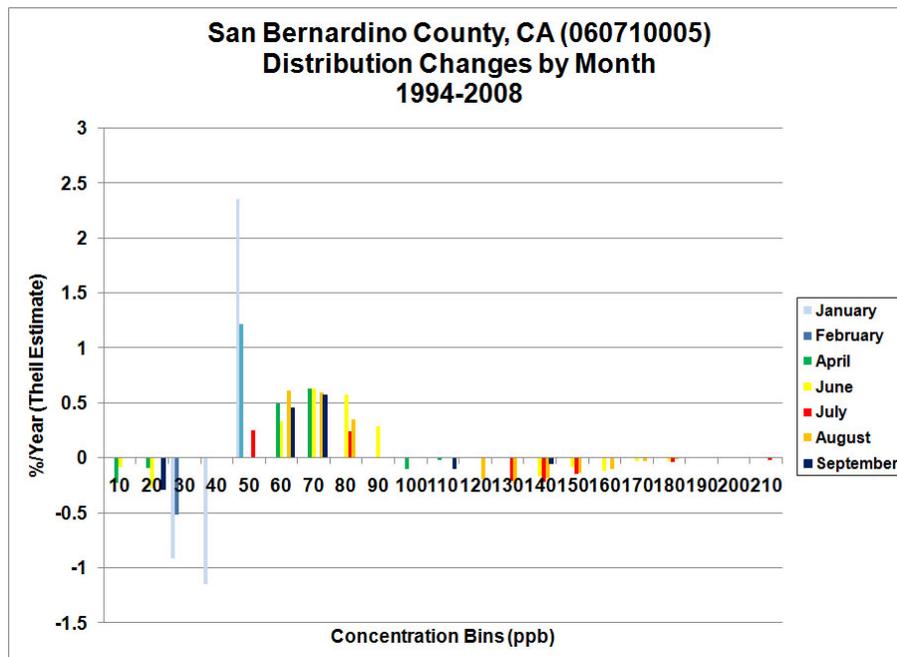


Fig. E2. Distribution of changes by month for a monitoring site located in San Bernardino County, California (AQS 060710005) for 1994-2008 for the months with statistically significant changes. (Source: Lefohn et al., 2010).

In summarizing its mortality and morbidity risk results, on page 9-23 of the REA, EPA notes that the risks did not show large responses to meeting existing or alternative levels of the standard for several reasons.

- First, these risks were based on concentration-response (C-R) functions that were approximately linear along the full range of concentrations, and therefore reflected

the impact of changes in O₃ along the complete range of 8-hour average O₃ concentrations. This included days with *low baseline O₃ concentrations* that were predicted to have *increases* in O₃ concentrations, as well as days with *higher starting O₃ concentrations that were predicted to have decreases in O₃ concentrations* as a result of just meeting existing and alternative standards.

- Second, these risks, according to the EPA, reflected changes in the urban-area wide monitor average, which would not be as responsive to air quality adjustments as the design value monitor, and which included monitors with both decreases and increases in 8-hour concentrations.
- *Third, the days and locations with predicted increases in O₃ concentrations (generally those with low to midrange starting O₃ concentrations) resulting from just meeting the existing or alternative standard levels generally were frequent enough to offset days and locations with predicted decreases in O₃. The heat maps presented in Figures 7-2 and 7-3 in the REA demonstrated that just meeting progressively lower alternative standard levels narrowed the distribution of risk across the range of O₃ concentrations. In addition, the distribution of risk tended to be more centered on area-wide average concentrations in the range of 25 to 55 ppb after just meeting an alternative standard of 60 ppb. The focus of the epidemiological studies on urban case study area-wide average O₃ concentrations, and the lack of thresholds coupled with the linear nature of the C-R functions meant that in this analysis, the impact of a peak-based standard (which seeks to reduce peak concentrations regardless of effects on low or mean concentrations) on estimates of mortality and morbidity risks based on results of those studies was relatively small.*

As indicated in the third bullet, the distribution of risk tended to be centered in the 25-55 ppb range of 8-hour daily maximum concentrations after just meeting an alternative standard of 60 ppb. Further investigating the data in Fig. 7-B1 in the REA Appendix for Chapters 7-9 on page 7B-3, in most cases it appears that the greatest percentage of risk tended to also be in the 25-55 ppb range for recent conditions (2007), current standard (75 ppb), alternative standard (70 ppb), alternative standard (65 ppb), and alternative standard (60 ppb). Fig. E3 illustrates that reducing emissions to attain the various standards increased the risk in the 25-55 ppb mid-range concentrations from the current conditions and this range of concentrations made up the greatest percentage of the risk.

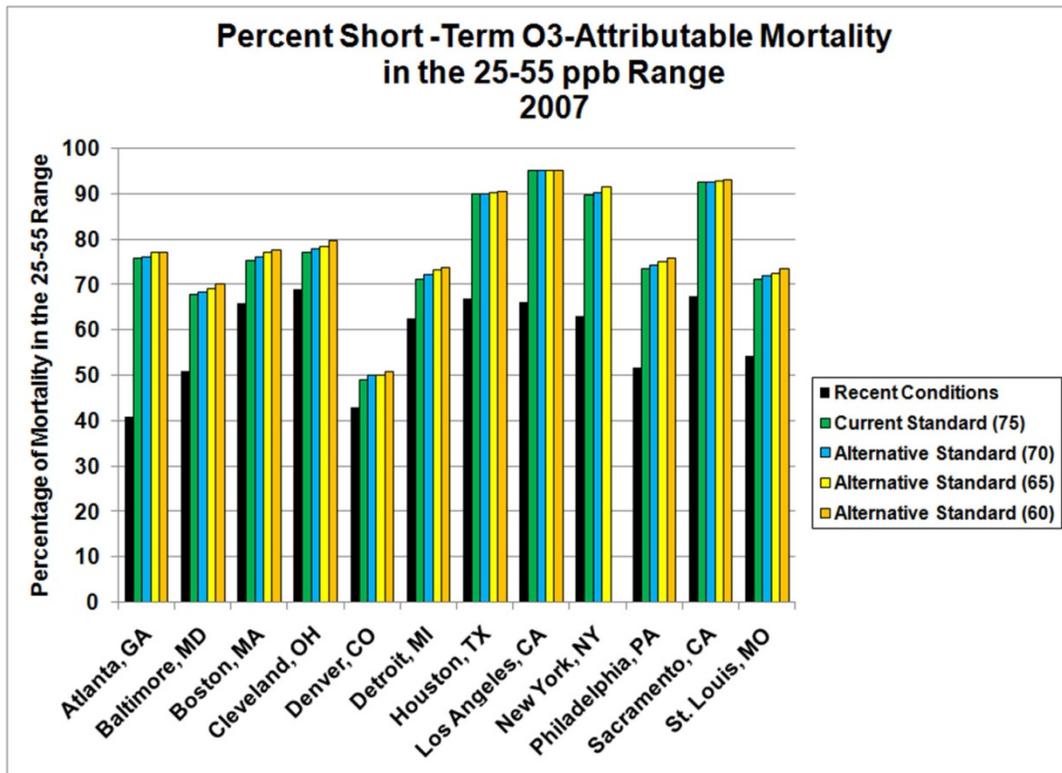


Fig. E3. Percent short-term O₃-attributable mortality in the 25-55 ppb range for various exposure conditions for 2007. (Source: Data from Fig. 7-B1 on page 7-B3 of the REA Appendix).

Compared to the mortality and morbidity risk assessments summarized above, as pointed out on page 9-44 of the REA, the lung function risk analysis is less sensitive to increases at the very low O₃ concentrations because the risk function is logistic and shows little response when ambient concentrations are generally less than 20 ppb for the 10 percent FEV1 decrement and generally less than 40 ppb for the 15 percent FEV1 decrement. For estimating the distribution of daily FEV1 decrements $\geq 10\%$ across ranges of 8-hour average ambient O₃ concentrations for cities and air quality scenarios, we have reviewed the distributions of composite monitor 8-hour daily maximum values for 12 urban case study areas in the epidemiology-based risk assessment (Fig. E4). The plots depict values based on ambient measurements (base), and values obtained with the HDDM adjustment methodology showing attainment of 75, 70, 65 and 60 ppb standards. Based on the distribution of concentrations shown in Fig. E4 reproduced from page 4-25 of the REA, we would anticipate as discussed in our comments that a large percentage of daily instances of FEV1 decrements $\geq 10\%$ would be predicted to occur when 8-hour average ambient concentrations were in the 25-55 ppb range for attainment of the 75, 70, 65 and 60 ppb standards.

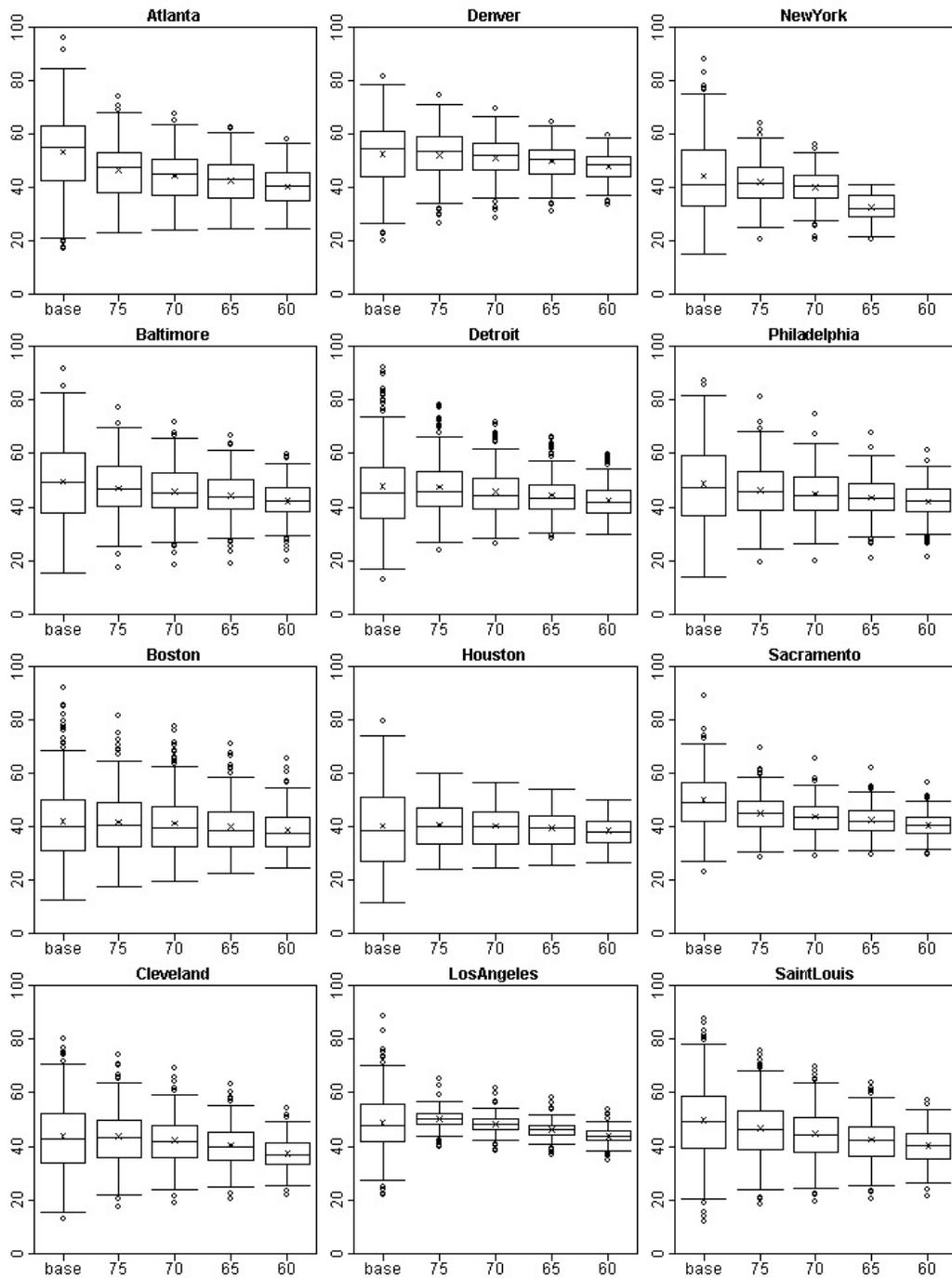


Fig. E4. Distributions of composite monitor 8-hour daily maximum values for the 12 urban case study areas in the epidemiology-based risk assessment. Plots depict values based on ambient measurements (base), and values obtained with the HDDM adjustment methodology showing attainment of 75, 70, 65 and 60 ppb standards. Values shown are based on CBSAs for April-October of 2007. Note that the HDDM 8 adjustment technique was not able to adjust air quality to show attainment of a 60 ppb standard in New York, so no boxplot is shown for that case. (Source: Fig. 4-9 of REA page 4-25).

The mid-range concentrations of 25-55 ppb have been highlighted both in the EPA's REA and in our comments. We have noted that the mid-level range of concentrations has an important effect on the estimated epidemiological and lung function risks. As anthropogenic emissions are reduced, increases in risks occur with the result that a large percentage of the risks are accumulated in the 25-55 ppb range of concentrations. In reviewing Fig. E5, which is presented in our comments, the contribution of hourly background O₃ to total observed O₃ in the 25-55 ppb range is large. Emissions-Influenced Background (EIB) O₃ as presented in the figures represents “titrated” background O₃ by anthropogenic sources. For example, at Yellowstone NP, the contribution of background O₃ in this range of concentrations is greater than 80% of total O₃; at Denver, background O₃ generally contributes between 70-80% to total O₃ in the 25-55 ppb range; and at Atlanta, EIB O₃ contributes to total O₃ approximately 50-70% in this range of concentrations.

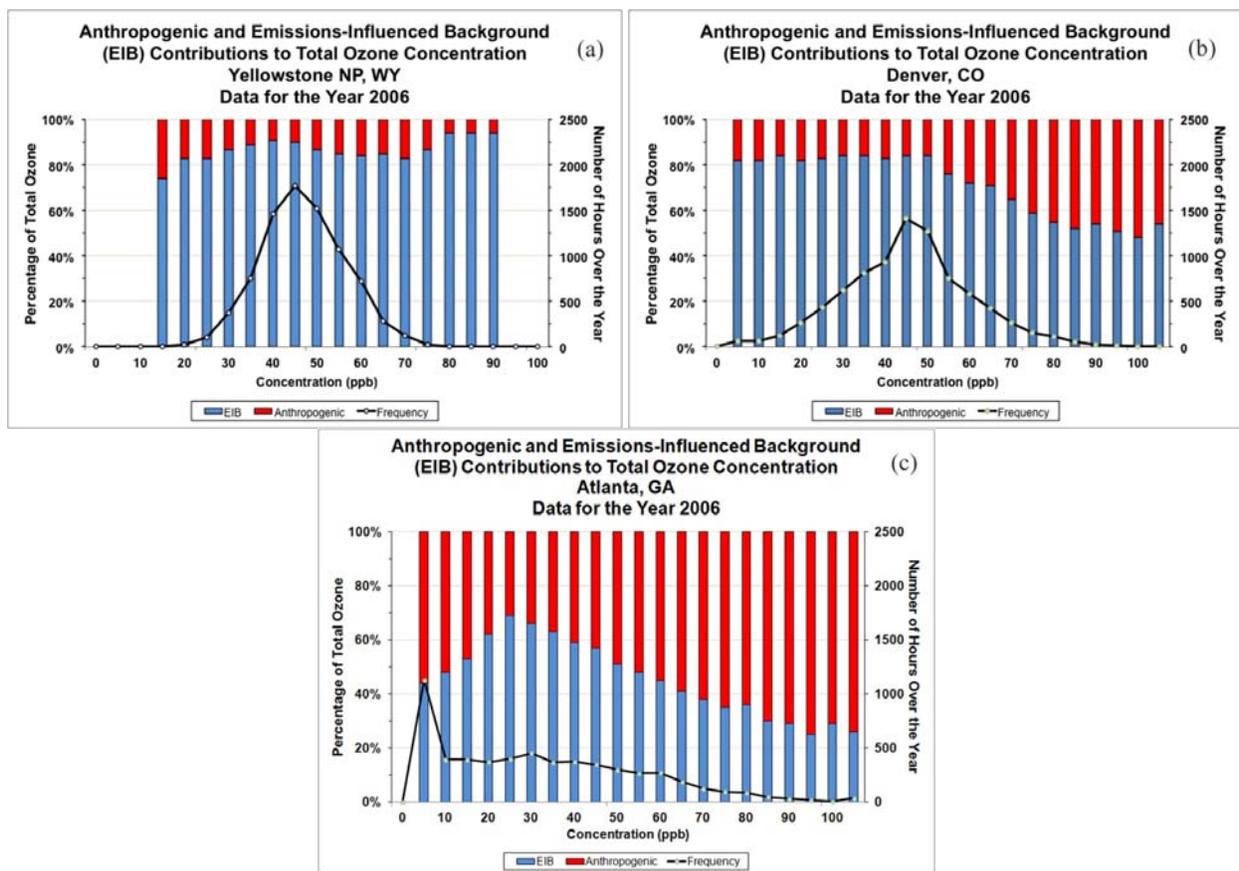


Fig. E5. Binned (5 ppb) frequency distribution of observed hourly total O₃ (black curve; right axis) and average relative binned contributions of maximum hourly EIB and anthropogenic O₃ (bars; left axis) for a) Yellowstone NP, b) Denver, and c) Atlanta. (Source: Lefohn et al., 2014a).

In the analysis of background O₃ we present in our comments, we find that that background O₃ was generally in the range of 30-70 ppb for the high-elevation sites and 30-45 ppb for the low-elevation site background site at Trinidad Head (CA). A similar range of background O₃ concentrations was observed for other low-elevation sites. For estimating

background O₃, we used results from the GEOS-Chem/CAMx (Lefohn et al., 2014a) and AM3 models (Lefohn et al., 2014b). As noted in our comments, for estimating values of EIB O₃ and NAB O₃, adjustments were required as described in Lefohn et al. (2014a, b) to account for likely underestimates (GEOS-Chem/CAMx) and overestimates (AM3) of background O₃ (as noted by Fiore et al., 2014).

We believe that the Agency has provided information that allows one to assess the relative contribution of (1) anthropogenic (i.e., controllable) O₃ and (2) background O₃ (non controllable) to its human health risk estimates. As noted in the REA, the lung function and epidemiological risk estimates for attaining the 75, 70, 65 and 60 ppb standards indicate that a large percentage of the risks are associated with 8-hour average ambient concentrations in the 25-55 ppb range, which is the range of concentrations associated with background O₃ and these concentrations are not controllable.

1. EPA's Approach in the REA

In the second draft of the REA (US EPA, 2014a) on page 3-12, the EPA, in consultation with CASAC, states that background O₃ concentrations can be ignored when estimating health risks because:

In the previous review, background O₃ (referred to in that review as policy relevant background, or PRB) was incorporated into the REA by calculating risk only in excess of PRB. CASAC members recommended that EPA move away from using PRB in calculating risks (Henderson, 2007). In addition, comments received from CASAC, based on their review of the first draft Risk and Exposure Assessment on September 11-12, 2012 (Frey and Samet, 2012), agreed with the development of risk estimates with reference to zero O₃ concentration. Based on these recommendations and comments, the second draft REA includes risks associated with O₃ from all sources after we have simulated just meeting the existing standard and estimates of total risk remaining after meeting alternative levels of the standards. EPA believes that presenting total risk is most relevant given that individuals and populations are exposed to total O₃ from all sources, and risks associated with O₃ exposure are due to total O₃ exposure and do not vary for O₃ exposure associated with any specific source. **In addition, background O₃ is fully represented in estimates of total risk given that the measured and adjusted air quality concentrations being used in the risk and exposure analyses include O₃ produced from precursor emissions from both anthropogenic and background sources** (emphasis added).

The EPA, while minimizing its discussion of background in the REA, states in the document that background O₃ will be discussed in the Policy Assessment (PA) report (EPA, 2014b). Although the EPA did not characterize background O₃ in the REA, the Agency actually has placed into perspective for CASAC, policy makers, and the general public in the REA how the relative contribution of (1) anthropogenic (i.e., controllable) O₃ and (2) background O₃ (non controllable) affects human health risk estimates.

As mentioned above by the EPA, the distribution of total O₃ consists of both anthropogenic and background contributions. Background O₃ distributions contribute to the risk estimates more and more as emissions are reduced to attain the various levels of existing and alternative O₃ standards. This is because as more stringent O₃ levels are attained, O₃

concentrations associated with anthropogenic emissions contribute less of a percentage to total O₃ than background O₃ concentrations. In the REA, the EPA estimates human health risks (attributable to both anthropogenic and background O₃) associated with lung function decrements and epidemiological endpoints down to 0 ppb for several air quality scenarios (i.e., at current O₃ conditions, at the existing O₃ primary standard level, and at various selected alternative O₃ primary standard levels). For the lung function decrements, the exposure-response function minimizes the contribution of the lower hourly average O₃ concentrations to risk. A series of modeling exercises is used to estimate the distribution of total O₃ concentrations (i.e., attributable to anthropogenic and background) that result from reducing emissions that are required to just meet the existing and alternative O₃ standards. In the REA on page 3-11, the EPA describes the process as follows:

Simulation of just meeting the existing and alternative O₃ standards is accomplished by adjusting hourly O₃ concentrations measured over the O₃ season using a model-based adjustment methodology that estimates O₃ sensitivities to precursor emissions changes. These sensitivities, which estimate the response of O₃ concentrations to reductions in anthropogenic NO_x and VOC emissions, are developed using the Higher-order Decoupled Direct Method (HDDM) capabilities in the Community Multi-scale Air Quality (CMAQ) model. This modeling approach incorporates all known emissions, including sources of natural and anthropogenic emissions in and outside of the U.S. By using the model-based adjustment methodology we are able to more realistically simulate the temporal and spatial patterns of O₃ response to precursor emissions. We chose to simulate just meeting the existing and alternative standards in the urban cast study areas by decreasing U.S. anthropogenic emissions of NO_x and VOC throughout the U.S. using equal proportional decreases in emissions throughout the U.S., in order to avoid any suggestion that we are approximating a specific emissions control strategy that a state or urban area might choose to meet a standard.

As will be discussed in the sections that follow, background O₃ is important at current concentrations and plays an important role in the risk estimates characterized by the EPA in the REA.

2. Human Health Risk Outcomes

2.1 The Percent Contribution of Background O₃ to Total Observed O₃

Background O₃ is of interest to researchers for the following two reasons: (1) at times background O₃ is associated with high concentrations (e.g., exceptional events in the Intermountain West) and (2) background O₃ contributes on a continuous basis to the distribution of observed hourly average O₃ concentrations and affects risk estimates. Background O₃ concentrations vary by the hour and are generally highest during the springtime at many O₃ monitoring sites across the US (see Lefohn et al. [2014a] for a discussion of emission-influenced background or referred to as apportionment-based US background in the PA). Fig. 1 illustrates the monitored 8-hour O₃ design values across the US for 2006-2008 (source: page 4-3 of REA). The highest O₃ concentrations are experienced in the southern California area with the next highest exposures occurring in the Eastern US. Fig. 2 shows the apportionment-based US background percent contribution to seasonal mean O₃ based on 2007 CAMx source apportionment modeling (source: page 2-18 of PA). As indicated in Fig. 2, a large percentage (i.e., >50%) of the 8-hour concentrations measured across the US consists of background O₃. In the West, Intermountain West, and the Northeast, the percentage contribution of background O₃ to the seasonal mean 8-hour concentration is 70% or greater.

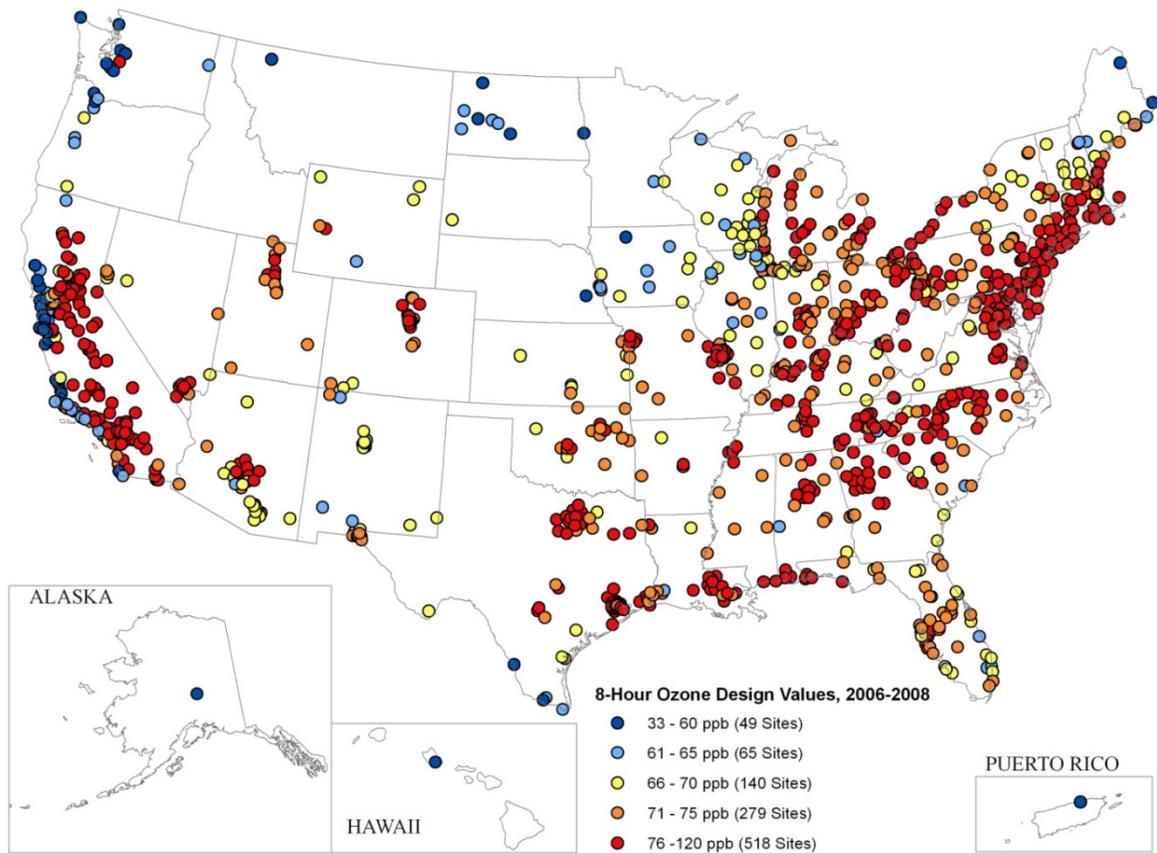


Fig. 1. The monitored 8-hour O₃ design values across the US for 2006-2008. (Source: page 4-3 of REA).

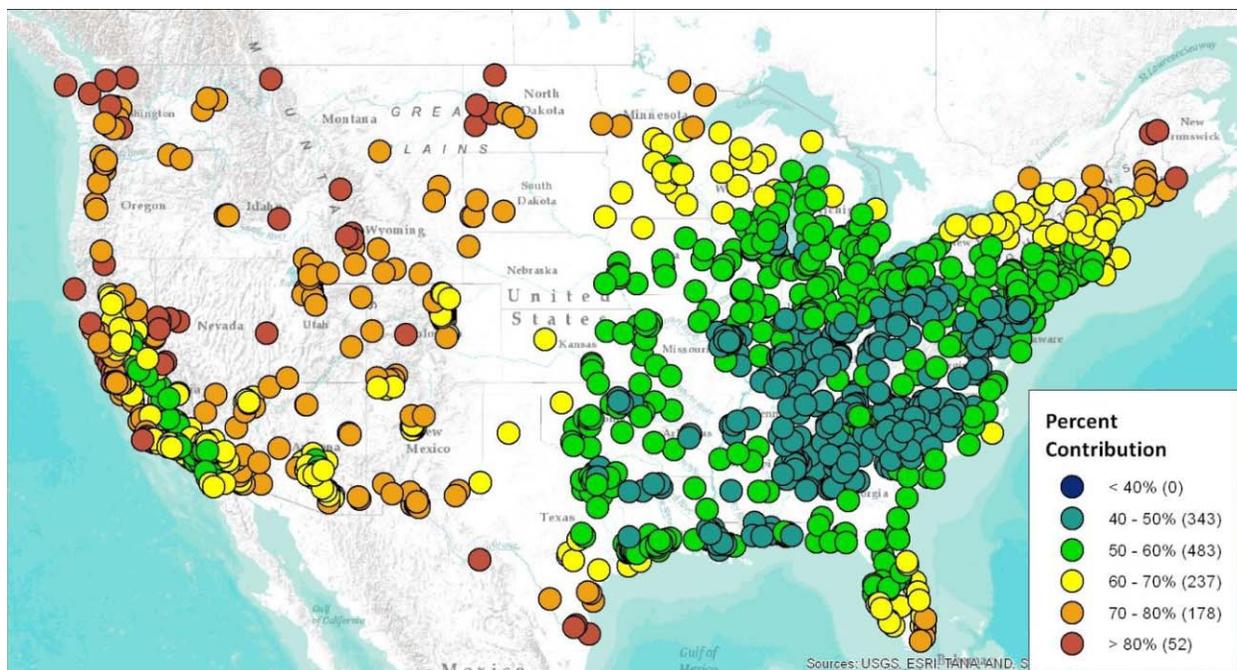


Fig. 2. Map of apportionment-based U.S. background percent contribution to seasonal mean O_3 based on 2007 CAMx source apportionment modeling. (Source: page 2-18 of PA).

The EPA results agree in general with the percent contributions of background O_3 estimated by Lefohn et al. (2014a) in their recent publication in *Atmospheric Environment*, which provided the characterization of background O_3 for 23 urban- and rural-influenced sites across the US. Fig. 3 illustrates the binned (5 ppb) frequency distribution of observed hourly total O_3 and average relative binned contributions of maximum hourly background O_3 (i.e., emission-influenced background) and anthropogenic O_3 for a) Yellowstone NP, b) Denver, and c) Atlanta.

Emission-influenced background (EIB) O_3 , as described by Lefohn et al. (2014a), is similar to the definition of apportionment-based US background as defined by the EPA on page 2-16 of the PA. EIB, as described in Lefohn et al. (2014a), is apportioned to global tropospheric O_3 and stratospheric O_3 entering North America and natural O_3 , formed in North America from continental biogenic, fire, and lightning sources. The simulation of all three EIB components includes chemical decay via interactions with North American anthropogenic and natural

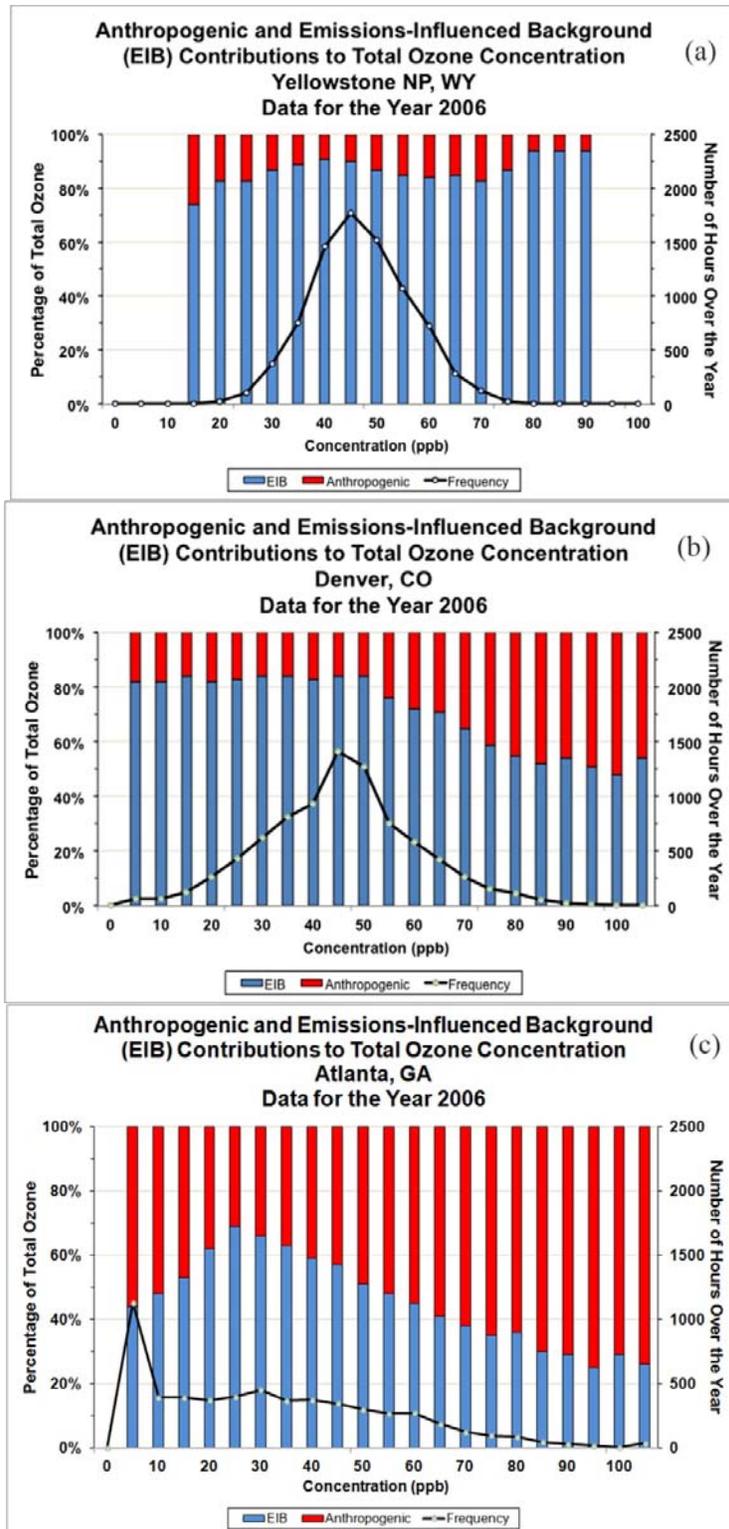


Fig. 3. Binned (5 ppb) frequency distribution of observed hourly total O₃ (black curve; right axis) and average relative binned contributions of maximum hourly EIB and anthropogenic O₃ (bars; left axis) for a) Yellowstone NP, b) Denver, and c) Atlanta. (Source: Lefohn et al., 2014a).

precursor emissions of nitrogen oxides (NO_x), volatile organic compounds (VOC), and carbon monoxide (CO). Unlike historical modeled background definitions that reflect the absence of anthropogenic emissions, EIB is defined to include chemical interactions with anthropogenic emissions; thus reducing O₃ lifetime in the polluted boundary layer and reflecting “current” background levels at the sites analyzed. Similarly, for apportionment-based US background, EPA estimates source apportionment model estimates of O₃ that is *attributable to sources other than U.S. anthropogenic emissions*. EPA notes that the advantage of the source apportionment modeling is that all of the modeled O₃ is attributed to various source terms and thus the Agency's approach is not influenced by the confounding occurrences of background O₃ values when zero-out modeling (i.e., anthropogenic emissions are eliminated in the US, North America, or everywhere) is implemented. The distinction between EIB (and apportionment-based US background) and EPA's hypothetical North American Background (NAB) O₃ and US Background O₃ (NAB) is that NAB and USB represent background O₃ concentrations that might be attained if anthropogenic emissions were reduced. In contrast, both EIB O₃ and apportionment-based US Background O₃ are estimated to represent levels of the "titrated" hypothetical NAB O₃ or USB O₃. In pristine areas with small anthropogenic influences, EIB O₃ is similar to NAB O₃. In urban areas, EIB O₃ is chemically decayed but converges upward toward the higher NAB O₃ level as anthropogenic emissions are reduced. As anthropogenic emissions are reduced in North America, EIB O₃ approaches NAB O₃. Similarly, as anthropogenic emissions are reduced in the US, apportionment-based US Background O₃ approaches USB O₃. In other words, EIB O₃ is less than or equal to NAB O₃; similarly, apportionment-based US Background O₃ is less than or equal to USB O₃ depending upon the degree of influence of anthropogenic emissions.

Fig. 3 plots the modeled contributions of hourly background O₃ (i.e., EIB) to the total O₃ frequency distribution (black curve) over the entirety of 2006. The estimated EIB O₃ at Yellowstone NP contributes > 75% to total O₃ at each part of the distribution (Fig. 3a); at Denver the EIB O₃ contributes > 50% to total O₃ at each part of the distribution (Fig. 3b) and at Atlanta, the EIB O₃ contributes > 50% of mid-range total O₃ (30-50 ppb) with a lower contribution outside this range as result of dominant local anthropogenic influences (Fig. 3c). While the large percentage of background O₃ (i.e., EIB O₃) for Yellowstone NP at the higher concentrations is associated with stratospheric tropospheric transport processes (i.e., episodic natural occurrences), the concentrations at both the mid- and low-range concentration levels are heavily influenced by global tropospheric O₃, with important contributions (i.e., enhancements) from the stratosphere (Lin et al., 2012; Lefohn et al., 2014a). In contrast to Yellowstone NP, the higher O₃ concentrations experienced at Denver and Atlanta are influenced by anthropogenic sources. The percentages presented in Fig. 3 from Lefohn et al. (2014a) for the three locations are similar to those estimated by the EPA in Fig. 2. The Appendix contains additional figures similar to Fig. 3 for an additional 20 locations. The percentages in those figures are similar to the EPA estimates in Fig. 2.

As summarized earlier, EPA has made the decision to represent total risk independent of any specific source (i.e., anthropogenic and background). This decision results in estimated risks that are associated with both controllable and uncontrollable sources (i.e., natural). As observed in Fig. 2 (from EPA's PA) and Fig. 3 (from Lefohn et al., 2014a) above, background O₃ makes up a large percentage of the monitored observed O₃ concentrations and therefore, as we will see, plays an important role in comparing the REA risk outcomes across base conditions, attaining the current standard, and attaining alternative standard levels.

While several researchers (e.g., Zhang et al., 2013; Fiore et al., 2014) suggest that an important use of models (e.g., GEOS-Chem and AM3) is to assist the EPA in identifying exceptional O₃ events in the West and Intermountain West when the O₃ NAAQS is exceeded, we see a larger role for models in characterizing background O₃ levels and how these levels affect EPA's REA risk outcomes and the attainability of the O₃ NAAQS. While important differences in the magnitude and spatial and temporal variability of background O₃ estimates exist among the various models used to predict background O₃ (Zhang et al., 2013; Fiore et al., 2014; Lapina et al., 2014), the models can assist in assessing both human health risk estimates and the attainability of the NAAQS.

2.2 The Effects of O₃ Distribution Shifts on Risk Characterization Metrics

Risk characterization is the process of communicating the results of risk (and exposure) modeling in terms (i.e., metrics) that decision makers can understand (page 2-23 of the REA). In the REA, EPA notes that this translates into providing metrics that are most useful in the Policy Assessment to assess the adequacy of the existing O₃ standards in protecting public health with an adequate margin of safety and to evaluate the additional protection provided by potential alternative standards. The EPA notes in the REA and the Policy Assessment that the Agency has selected aggregate risk metrics, including the number and percent of vulnerable populations experiencing adverse respiratory responses based on application of results of controlled human exposure studies and the attributable incidence and percent of baseline incidence of mortality and morbidity endpoints based on application of results of epidemiology studies. For all three types of metrics (i.e., exposure, risk based on controlled human exposure studies, and risk based on epidemiology studies) and for the purpose of evaluating the adequacy of the existing standards,

the focus in the REA is on the exposure and risk remaining upon just meeting the existing O₃ standards.

As noted by the EPA in the REA (page 9-31) in summarizing its risk results, many of the differences in the risk results across the metrics used to quantify risk were driven by how each metric was affected by the O₃ data input to the individual analysis. EPA noted that in general, the impact of the HDDM adjustments to O₃ varied based on three main considerations: 1) the degree to which the exposure or risk metric was sensitive to changes across the various ranges of O₃ concentrations (e.g., high, mid-range, low); 2) whether the exposure or risk metric used individual census tract concentrations or area-wide average concentrations; and 3) changes in the distribution of O₃ concentrations in the year of analysis between recent O₃ concentrations and adjusted (meeting the existing or alternative standards) O₃ scenarios.

One of the important key elements associated with the risk estimates is the change in the distribution of O₃ concentrations between recent O₃ concentrations and adjusted (meeting the existing or alternative standards O₃ scenarios). With respect to the changes in the distribution of O₃ concentrations as a function of emission reductions, the risk metrics used in the REA were influenced by how the distribution of O₃ concentrations change (REA, page 9-32). The change in the distribution of hourly average O₃ concentrations results from emission reductions. These emissions reductions change the distribution of O₃ concentrations in the high-, mid-, and low-range values. A way to provide information on the pattern of changes is by characterizing the distribution of hourly average concentrations as higher hourly average O₃ concentrations are reduced as a result of lowering NO_x emissions.

How distributions of O₃ concentrations change as emissions are reduced has been documented in the literature. Lefohn et al. (1998) noted that as O₃ levels improved (i.e., the

environment experienced lower O₃ exposures) due to reduced emissions, reductions in the number of high hourly average concentrations, as well in the number of low hourly average concentrations occurred. The reduction in the number of low hourly average O₃ concentrations was associated with lack of NO_x scavenging (US EPA, 1996). Lefohn et al. (1998) predicted that as a site's air quality improved, the distribution of the hourly average concentrations would move from both the high end as well as the low end of the distribution toward the center (i.e., 30-60 ppb). The authors illustrated the frequency of occurrences of hourly average concentrations for a heavily urban-influenced monitoring site at Jefferson County, Kentucky (Fig. 4a). The urban-influenced site in Kentucky showed frequent high and low hourly average concentrations. This site appeared to be influenced by NO_x scavenging because of the occurrence of frequent low hourly average concentrations. In contrast to this site, Lefohn et al. (1998) showed the distribution pattern for a rural site in the Custer National Forest in Montana (Fig. 4b) that experienced very low maximum hourly average concentrations. The distribution of the hourly average concentrations at the Montana site showed a lack of both high and low hourly average concentrations due to a lack of NO_x scavenging. The Montana site experienced its highest hourly average concentrations in the April-May period, similar to sites in the Intermountain West. Lefohn et al. (1998) hypothesized that as anthropogenic emissions were reduced for heavily urban-influenced sites, the hourly average concentration distribution patterns similar to the Jefferson County, KY site would shift from both the high end as well as the low end of the distribution toward the mid-range concentrations (i.e., 30-60 ppb) and the distribution would ultimately look similar to the site. Several investigators (Oltmans et al., 2006; Lefohn et al., 2008; Oltmans et al., 2008; Lefohn et al., 2010; Oltmans et al., 2013) have reported trending results that showed shifts in the distribution over time, where both the high and the low ends of

the distributions of O₃ concentrations shifted toward the mid-level values. Lefohn et al. (2010) illustrated the concentrations shifts by month for a site in San Bernardino (CA) for the period 1994-2008 (Fig. 5).

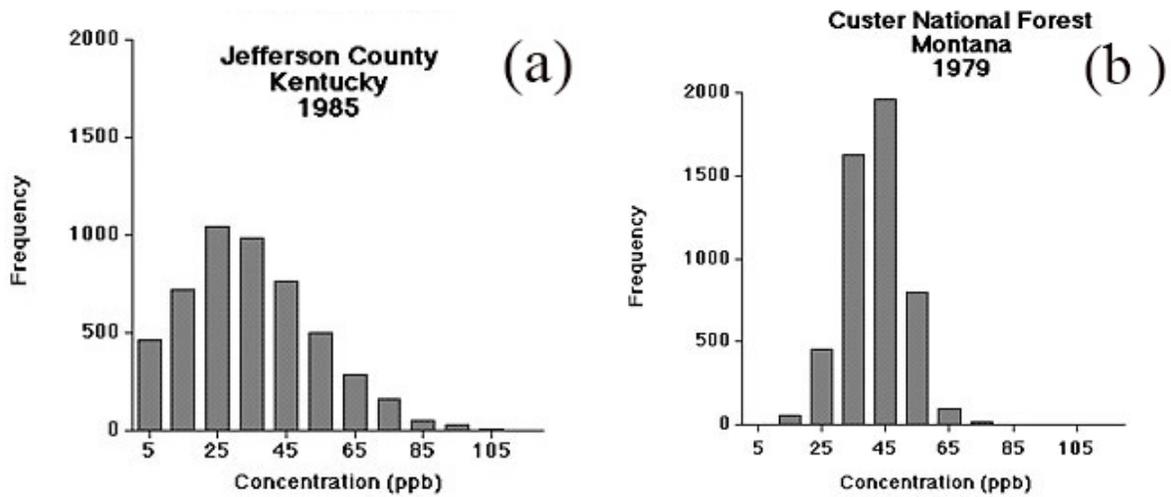


Fig. 4. A comparison of the distribution of hourly average O₃ concentrations site at a (a) heavily urban-influenced Jefferson County (KY) and (b) rural site at Custer National Forest (MT). (Source: Lefohn et al., 1998).

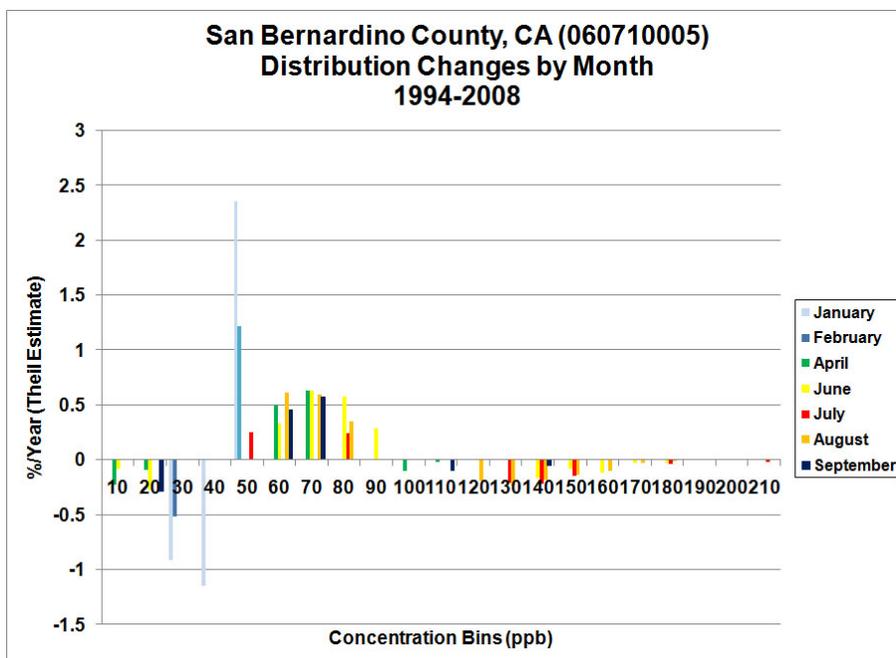


Fig. 5. Distribution of changes by month for a monitoring site located in San Bernardino County, California (AQS 060710005) for 1994-2008 for the months with statistically significant changes. (Source: Lefohn et al., 2010).

As noted above, the change in the distribution of O_3 concentrations results from emission reductions. These shifts in distribution affect both estimated lung function and epidemiological risks. *On page 9-23 of the REA, EPA summarized its mortality and morbidity risks by observing that generally these risks did not show large responses to meeting existing or alternative levels of the standard for several reasons.*

- First, these risks were based on concentration-response (C-R) functions that were approximately linear along the full range of concentrations, and therefore reflected the impact of changes in O_3 along the complete range of 8-hour average O_3 concentrations. This included days with *low baseline O_3 concentrations* that were predicted to have *increases* in O_3 concentrations, as well as days with *higher starting O_3 concentrations that were predicted to have decreases in O_3 concentrations* as a result of just meeting existing and alternative standards.
- Second, these risks, according to the EPA, reflected changes in the urban-area wide monitor average, which would not be as responsive to air quality adjustments as the design value monitor, and which included monitors with both decreases and increases in 8-hour concentrations.

- ***Third, the days and locations with predicted increases in O₃ concentrations (generally those with low to midrange starting O₃ concentrations) resulting from just meeting the existing or alternative standard levels generally were frequent enough to offset days and locations with predicted decreases in O₃. The heat maps presented in Figures 7-2 and 7-3 in the REA demonstrated that just meeting progressively lower alternative standard levels narrowed the distribution of risk across the range of O₃ concentrations. In addition, the distribution of risk tended to be more centered on area-wide average concentrations in the range of 25 to 55 ppb after just meeting an alternative standard of 60 ppb. The focus of the epidemiological studies on urban case study area-wide average O₃ concentrations, and the lack of thresholds coupled with the linear nature of the C-R functions meant that in this analysis, the impact of a peak-based standard (which seeks to reduce peak concentrations regardless of effects on low or mean concentrations) on estimates of mortality and morbidity risks based on results of those studies was relatively small.***

As indicated in the third bullet, the distribution of risk tended to be centered in the 25-55 ppb range of 8-hour daily maximum concentrations after just meeting an alternative standard of 60 ppb. Further investigating the data in Fig. 7-B1 in the REA Appendix for Chapters 7-9 on page 7B-3, in most cases it appears that the greatest percentage of risk tended to also be in the 25-55 ppb range for recent conditions (2007), current standard (75 ppb), alternative standard (70 ppb), alternative standard (65 ppb), and alternative standard (60 ppb). Fig. 6 also illustrates that reducing emissions to attain the various standards increased the risk in the 25-55 ppb mid-range concentrations from the current conditions and this range of concentrations made up the greatest percentage of the risk.

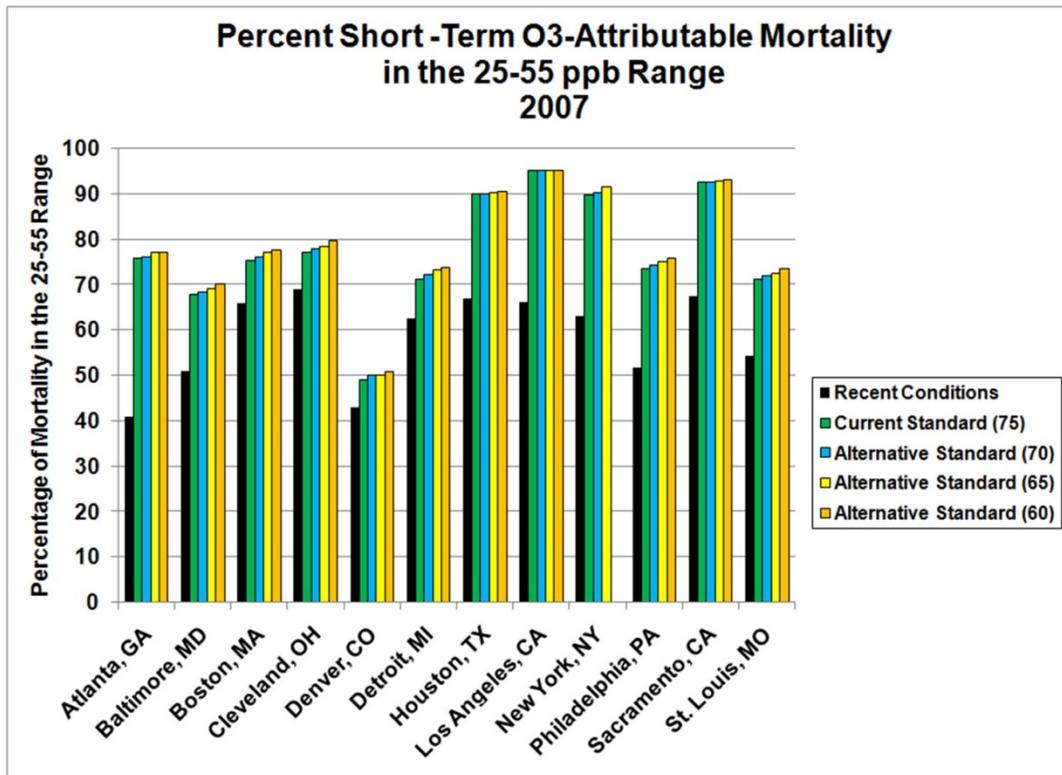


Fig. 6. Percent short-term O₃-attributable mortality in the 25-55 ppb range for various exposure conditions for 2007. (Source: Data from Fig. 7-B1 on page 7-B3 of the REA Appendix).

Compared to the mortality and morbidity risk assessments summarized above, as pointed out on page 9-44 of the REA, the lung function risk analysis is less sensitive to increases at the very low O₃ concentrations because the risk function is logistic and shows little response when ambient concentrations are generally less than 20 ppb for the 10 percent FEV1 decrement and generally less than 40 ppb for the 15 percent FEV1 decrement. Under current conditions in 2006,

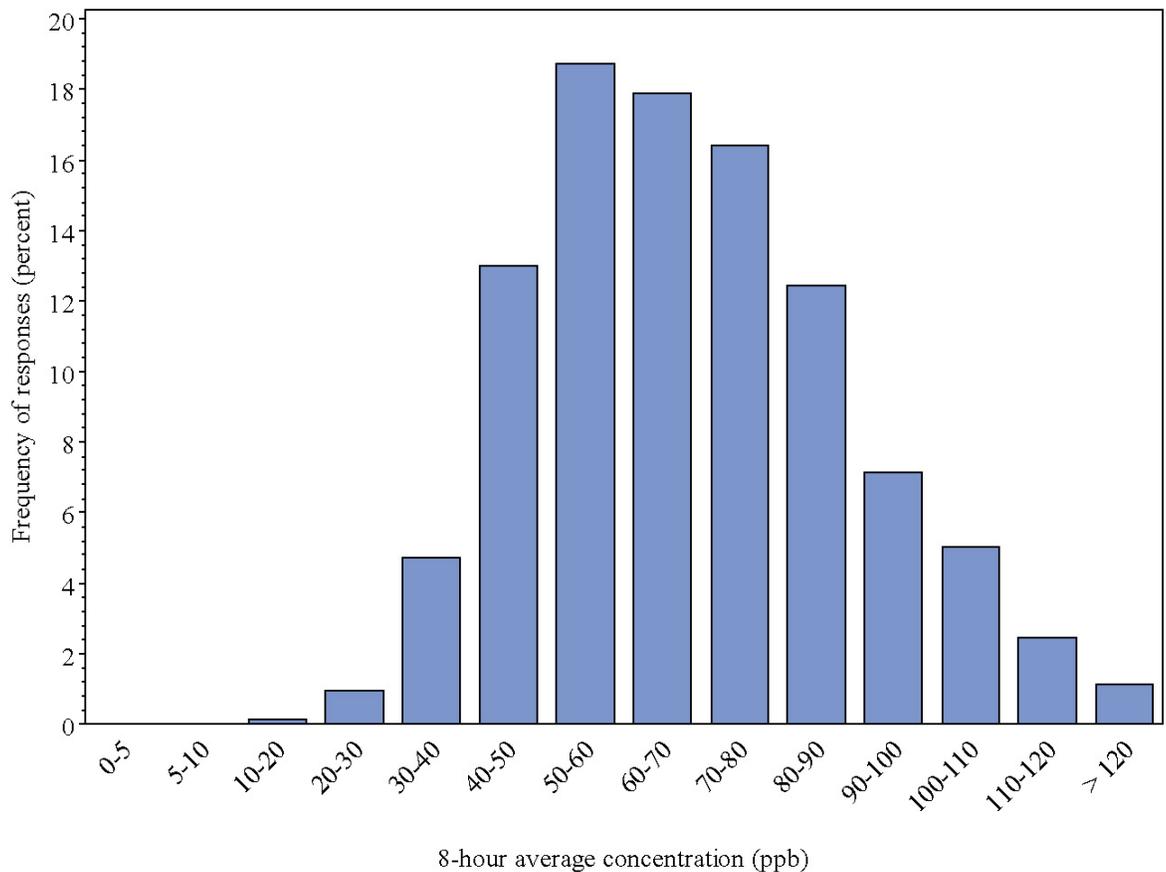


Fig. 7. Distribution of daily FEV1 decrements $\geq 10\%$ across ranges of 8-hour average ambient O_3 concentrations (Los Angeles, 2006 recent air quality). (Source: REA page 6-27).

Fig. 7 illustrates for Los Angeles that more than 90% of daily instances of FEV1 decrements $\geq 10\%$ occur when 8-hour average ambient concentrations are above 40 ppb for the modeled scenario. The distribution will be different than Los Angeles for different cities, years, and air quality scenarios. For estimating the distribution of daily FEV1 decrements $\geq 10\%$ across ranges of 8-hour average ambient O_3 concentrations for other cities and air quality scenarios, we have reviewed the distributions of composite monitor 8-hour daily maximum values for 12 urban case study areas (Fig. 8). The plots depict values based on ambient measurements (base), and values

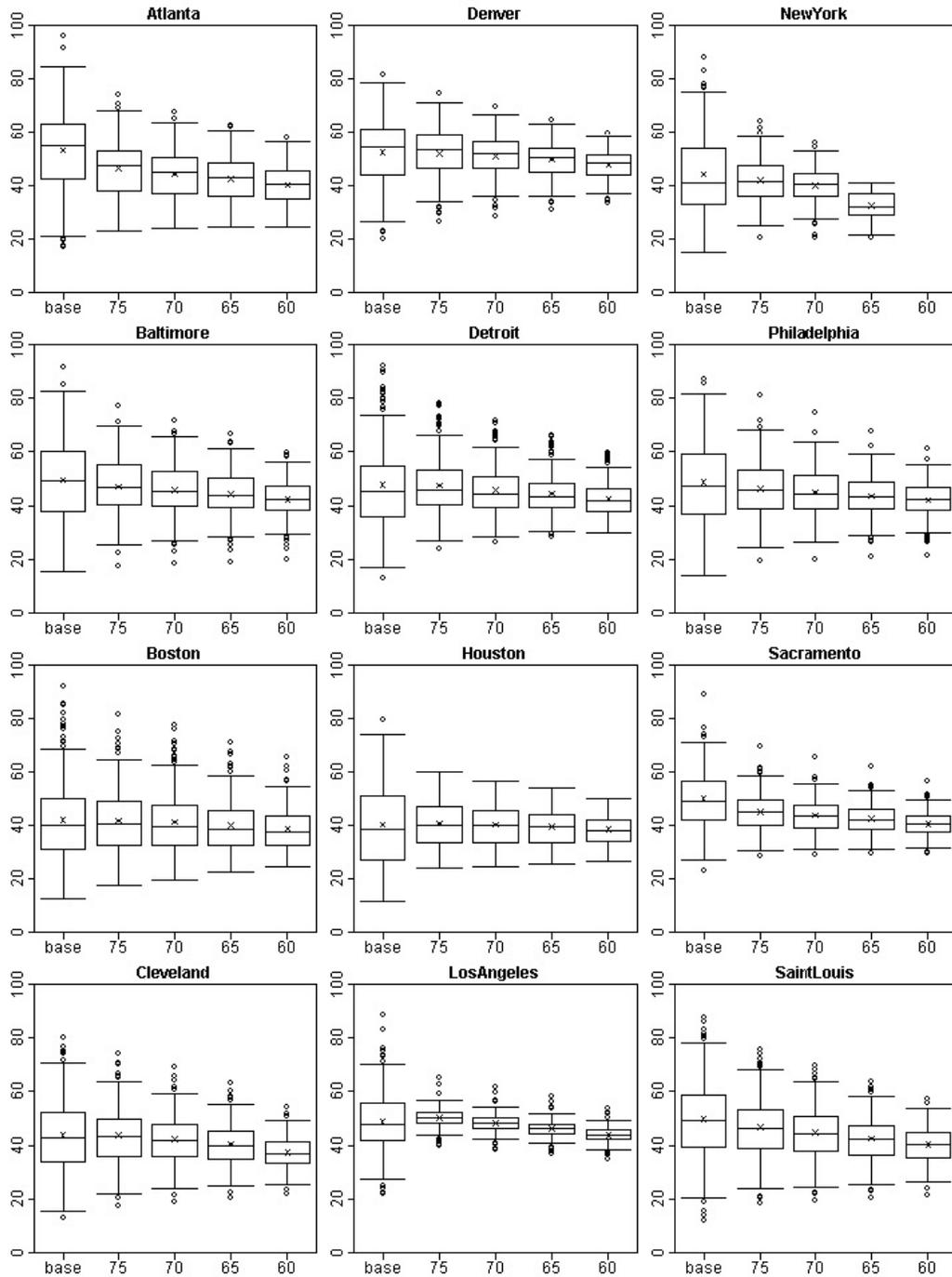


Fig. 8. Distributions of composite monitor 8-hour daily maximum values for the 12 urban case study areas in the epidemiology-based risk assessment. Plots depict values based on ambient measurements (base), and values obtained with the HDDM adjustment methodology showing attainment of 75, 70, 65 and 60 ppb standards. Values shown are based on CBSAs for April-October of 2007. Note that the HDDM 8 adjustment technique was not able to adjust air quality to show attainment of a 60 ppb standard in New York, so no boxplot is shown for that case. (Source: Fig. 4-9 of REA page 4-25).

obtained with the HDDM adjustment methodology showing attainment of 75, 70, 65 and 60 ppb standards. Based on the distribution of concentrations shown in Fig. 8 above, we would anticipate that a large percentage of daily instances of FEV1 decrements $\geq 10\%$ would be predicted to occur when 8-hour average ambient concentrations were in the 25-55 ppb range for attainment of the 75, 70, 65 and 60 ppb standards. The range of the distribution of daily FEV1 decrements $\geq 10\%$ across ranges of 8-hour average ambient O₃ concentrations is a function of the estimated ambient O₃ concentrations, exposure-response relationship, number of subjects exposed to specific ambient O₃ levels, and other various variables in the lung function model. We anticipate that increases in the mid-range concentrations (25-55 ppb), associated with reduced anthropogenic emissions, would result in a substantial percentage of predicted daily instances of FEV1 decrements $\geq 10\%$ occurring in this range as the 75, 70, 65 and 60 ppb standards were attained.

2.3 The Range of Background O₃ Concentrations

The mid-range concentrations of 25-55 ppb have been highlighted both in the EPA's REA and in our comments. We have noted that the mid-level range of concentrations has an important effect on the estimated epidemiological and lung function risks. As anthropogenic emissions are reduced, increases in risks occur with the result that a large percentage of the risks are accumulated in the 25-55 ppb range of concentrations. In reviewing Fig. 3, which we presented earlier in our comments, for ease of comparison we have reproduced the figure below. The contribution of hourly background O₃ (i.e., EIB) to total observed O₃ in the 25-55 ppb range is large. For example, at Yellowstone NP (Fig. 3a), the contribution of background O₃ in this range of concentrations is greater than 80% of total O₃; at Denver (Fig. 3b), EIB O₃ generally

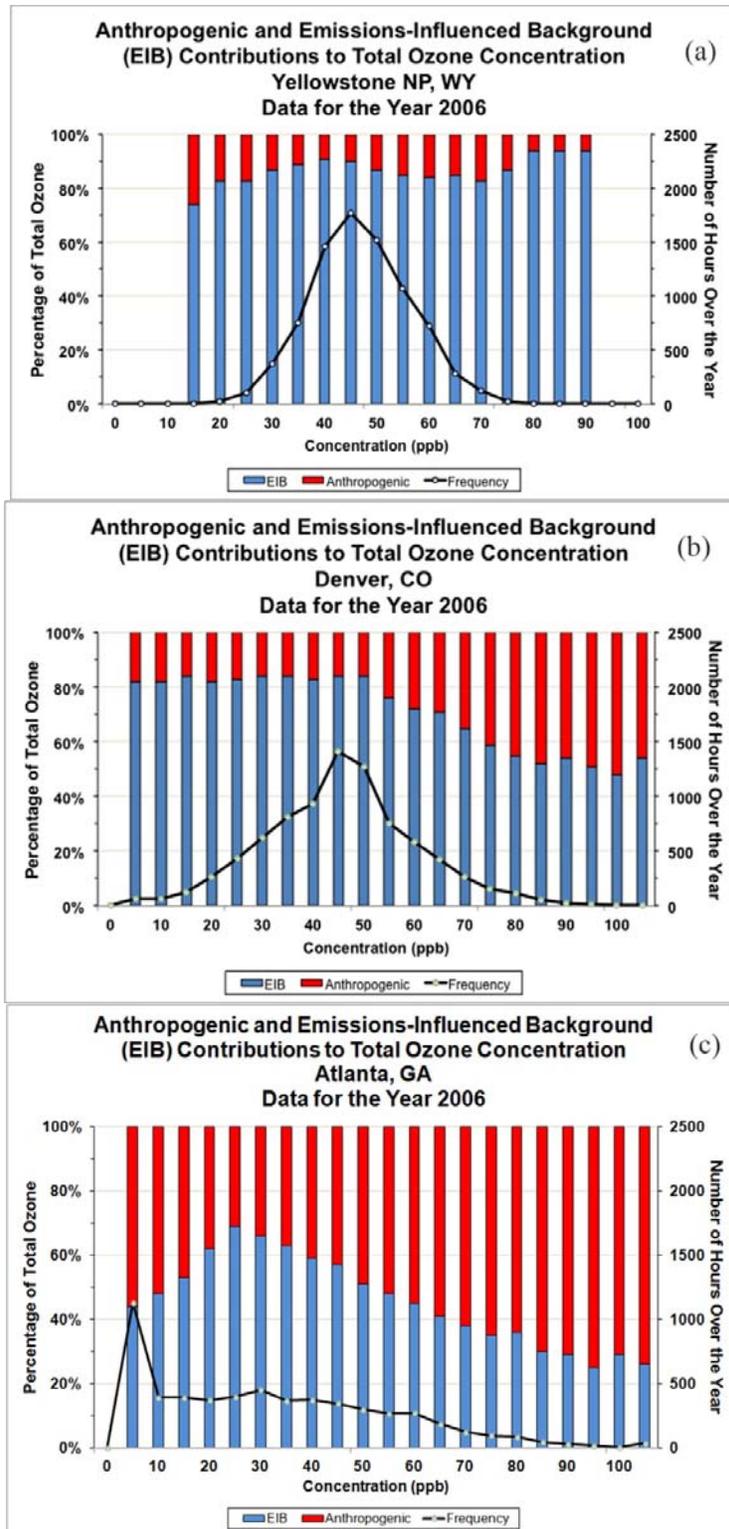


Fig. 3. Binned (5 ppb) frequency distribution of observed hourly total O₃ (black curve; right axis) and average relative binned contributions of maximum hourly EIB and anthropogenic O₃ (bars; left axis) for a) Yellowstone NP, b) Denver, and c) Atlanta. (Source: Lefohn et al., 2014a).

contributes between 70-80% to total O₃ in the 25-55 ppb range; and at Atlanta, EIB O₃ contributes to total O₃ approximately 50-70% in this range of concentrations. Similar important contributions of background O₃ in the 25-55 ppb range of concentrations are observed in the figures shown in the Appendix. Besides Yellowstone NP, Detroit, and Atlanta, results for Baltimore, Boston, Chicago, Cleveland, Dallas, Detroit, Houston, Los Angeles, New York, Philadelphia, Sacramento, Seattle, St. Louis, Washington DC, Georgia Station, Gothic, Pinedale, Shenandoah NP, Voyageurs NP, and Yosemite NP are presented.

As noted earlier, background O₃ as characterized by EIB O₃ in Fig. 3 as well as in the figures in the Appendix, represents "titrated" hypothetical NAB O₃. EIB O₃ is less than or equal to NAB O₃. In pristine areas with small anthropogenic influences, EIB O₃ is similar to NAB O₃. In urban areas, EIB O₃ is chemically decayed but converges upward toward the higher NAB O₃ level as anthropogenic emissions are reduced. Thus, as anthropogenic emissions are reduced to attain specific O₃ standard levels, background O₃ (as represented by EIB O₃) increases and approaches NAB O₃. Thus, the percentage contributions of background O₃ (i.e., EIB O₃) illustrated in Fig. 3 and the figures in the Appendix will increase in the 25-55 ppb range as emissions are reduced in the anthropogenically perturbed sites. The highest concentrations will disappear as well as the lower concentrations shown in the figures. Fig. 8, which we presented previously, illustrates the shifting from both ends of the concentration distribution toward the center as emissions are reduced.

Beside the shifting of the distribution of O₃ concentrations toward the mid-range values as emissions are reduced, the REA notes in Appendix 4D on pages 40-41 that while in most cities, the highest interquartile O₃ concentrations experienced in the recent condition scenario occur during the summer months (June-August), in many areas the highest interquartile O₃ concentrations shift to the spring months (i.e., April-May) for the adjustment scenarios of meeting 75 and 65 ppb

standard levels. This pattern would also occur if the analysis had included the 60 ppb scenario. The REA notes that the shifting seasonal pattern from summer to spring months is consistent with the higher contribution of non-US anthropogenic sources at lower standard levels than experienced under recent observed conditions. EPA notes on page 41 of Appendix 4D that many of these non-US anthropogenic sources are associated with stratospheric intrusions and international transport, which have been shown to peak during spring months (EPA, 2013).

Thus, based on the information provided in the REA, as well as in the published literature, as emission reductions of anthropogenic sources occur at urban-influenced sites, the following will be anticipated to occur:

- The highest concentrations experienced currently, as well as the lowest values of the distribution of concentrations experienced currently will be eliminated and the frequency of mid-level concentrations will increase;
- The percentage of background O₃ compared to total observed O₃ will increase in the mid-range concentrations of 25-55 ppb as emissions are reduced;
- As the frequency of mid-level concentrations increases as a result of emission reductions, based on the REA findings, the highest concentrations in the remaining O₃ distribution will occur during the springtime (i.e., April-May) versus the summertime (June-August);
- The contribution of non-US anthropogenic sources such as stratospheric intrusions and international transport, which peak during the spring months (EPA, 2013), will enhance estimated risk as well as contribute to potential NAAQS violations;
- As emissions are reduced, background O₃, which includes stratospheric intrusions and international transport, will increase its contribution to total O₃ at all O₃ monitoring sites, especially during the spring months; and
- The consequences of the 5 items above will be a more predominant contribution of background O₃ to both the epidemiological and lung function risk estimates characterized in the REA.

What do we know from the literature about the range of background O₃ concentrations?

Lefohn et al. (2014a) recently summarized the historical perspective of estimating background

O₃. While background O₃ cannot be measured directly, estimating it accurately is important. As we have discussed in our comments, background O₃ directly affect estimated human health risk and policy expectations regarding emission reduction effectiveness. McDonald-Buller et al. (2011) and EPA (2013) provide a summary of the concepts associated with background O₃ and its relevance to US air quality. Prior to 2006, O₃ measurements from remote monitoring sites were used to estimate background. EPA (1996) estimated hourly average summer background concentrations of 30-50 ppb and applied a background of 40 ppb in its risk analyses. EPA (2006) cited the work of Fiore et al. (2002, 2003), who applied the GEOS-Chem global model to estimate a mean background concentration range of 15-35 ppb. At that time, EPA (2006) defined North American background (NAB) O₃ to include contributions from global anthropogenic and natural sources in the absence of North American (i.e., U.S., Canada, Mexico) anthropogenic emissions. More recently, EPA (2013) has defined US background (USB) O₃ concentrations to include anthropogenic contributions from Canada and Mexico. Modeling results reported by EPA (2013) indicate USB and NAB concentrations tend to be higher in the West (particularly in the Intermountain West) and in the Southwest compared to the East in both spring and summer.

Lefohn et al. (2014a) describe the estimation of background O₃ over North America using the GEOS-Chem global model, the EPA Community Multi-scale Air Quality (CMAQ) regional model, the regional Comprehensive Air quality Model with extensions, and the AM3 global model. As noted in Lefohn et al. (2014a), each of these efforts reported incremental improvements, especially for the higher concentration ranges, by using greater resolution, updated modeling systems, and improved emissions and meteorological datasets. Modeling results (e.g., Zhang et al., 2011; Emery et al., 2012; Lin et al., 2012) estimate background O₃ ranges of 25-50 ppb across the US, with the highest peaks reaching well over 60 ppb in areas

affected by stratospheric intrusion and wildfires in the elevated areas of the western US. For the western US, results from Lin et al. (2012) illustrated the relative importance of stratospheric contributions to NAB. Lefohn et al. (2014a) noted that EIB O₃ contributes a major portion to estimated total O₃ for most sites in their analysis, especially during the spring. At the western high-elevation sites, the contributions of EIB to total O₃ were usually greater than 70% over the entire year. For many of the low-elevation sites, the contributions were 50% and higher during non-summer months. Patterns of higher spring EIB O₃ were followed by lower values during the summer, due to heightened chemical interaction with anthropogenic sources, which were then followed by rising EIB O₃ during the fall and winter months. For some high-elevation western US sites, this seasonal pattern was less discernible due to relatively small anthropogenic contributions and the high EIB O₃ estimated throughout the year.

Using data derived from Lefohn et al. (2014a), we characterized the MDA8 EIB O₃ time series for April, May, and June 2006 for Yellowstone NP, Pinedale, Gothic, and Yosemite NP (Fig. 9). The EIB O₃ concentrations represent “titrated” background O₃. For those sites that are influenced by anthropogenic emissions, EIB O₃ concentrations as represented in would increase as emissions were reduced. Fig. 9 Except for the periods when stratospheric O₃ intrusions influence the peak concentrations, the MDA O₃ values range generally between 40-60 ppb.

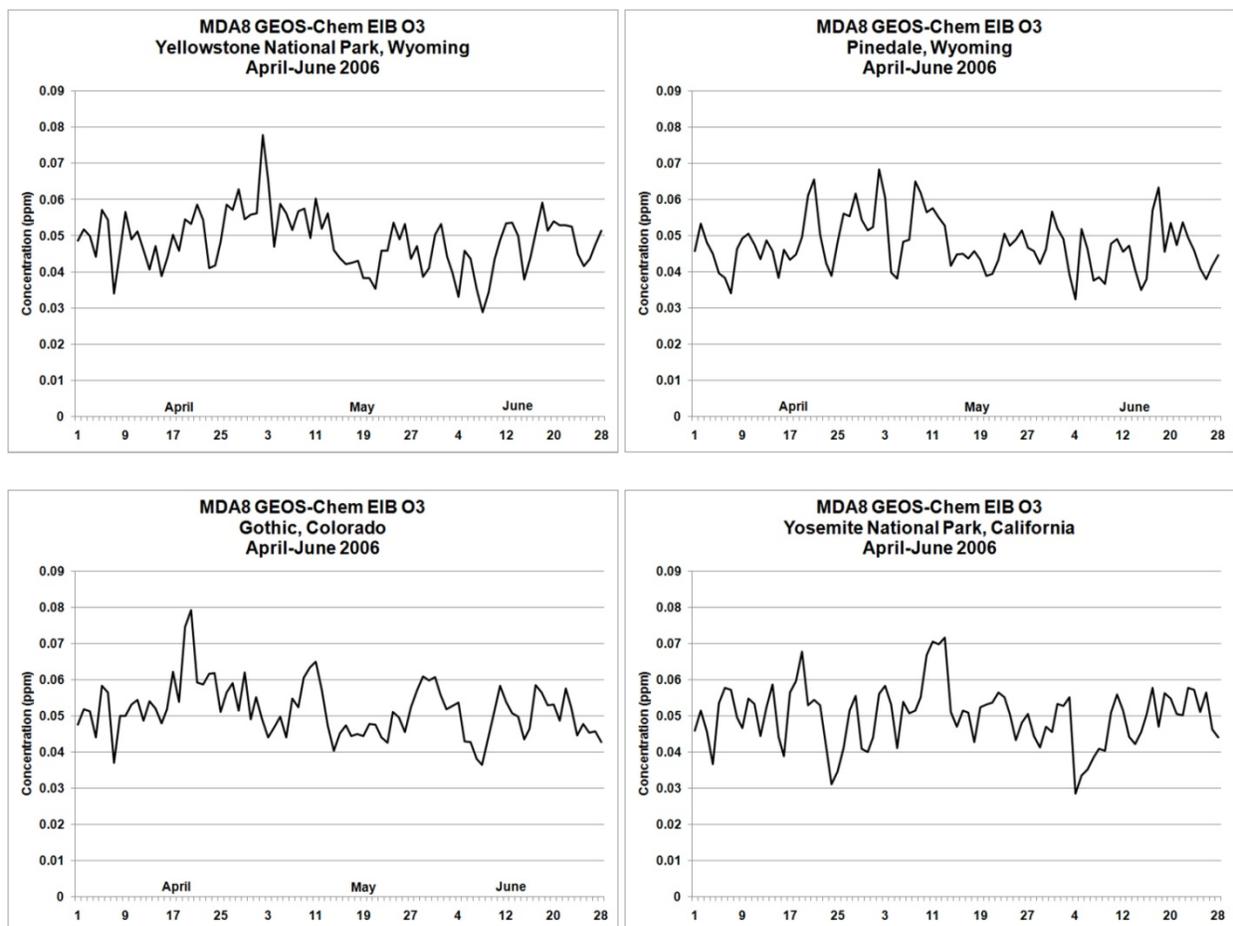


Fig. 9. MDA8 EIB O₃ time series for April-June 2006 for Yellowstone NP, Pinedale, Gothic, and Yosemite NP.

The high-elevation sites are influenced by global tropospheric O₃ (Lin et al., 2012; Lefohn et al., 2014a). Fig. 10 illustrates the MDA8 EIB O₃ time series for Denver and Sacramento. The EIB O₃ time series for the high-elevation site at Denver is similar to the sites in Fig. 9, which showed MDA O₃ values range generally between 40-60 ppb. The range of EIB O₃ concentrations for Sacramento is generally 20-50 ppb, which is a lower range of concentrations than for the other 5 sites. This reflects the Sacramento site's lower elevation as well as considerably more titration of background O₃ by anthropogenic emissions.

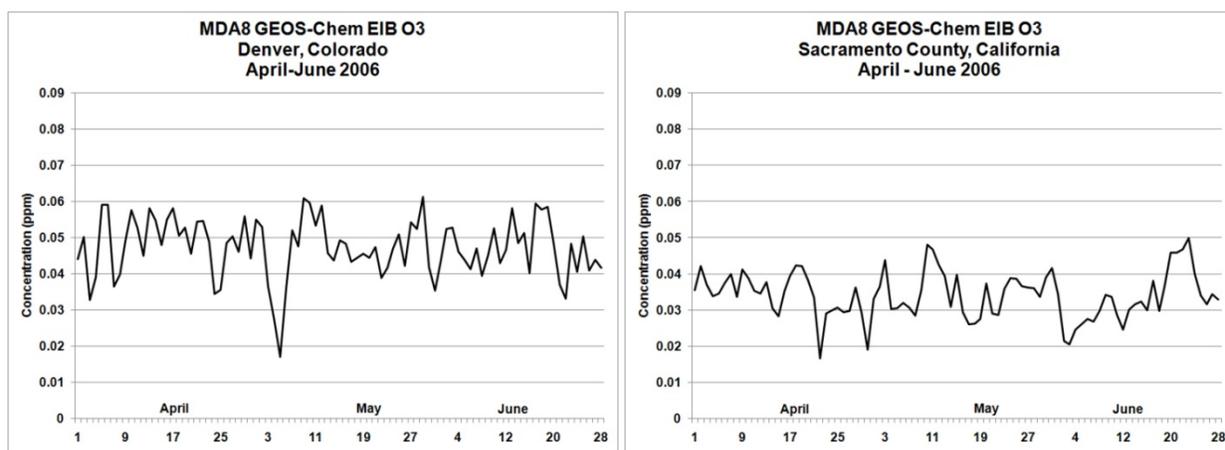


Fig. 10. MDA8 EIB O₃ time series for April-June 2006 for Denver and Sacramento.

Work by Lin et al. (2012) has estimated NAB in the spring based on high-resolution runs of the NOAA Geophysical Fluid Dynamics Laboratory AM3 model during the 2010 CALNEX campaign. A unique feature of the GFDL global coupled atmosphere-oceans-land-sea ice model within a general circulation model is a fully coupled stratosphere-troposphere chemistry component. Lefohn et al. (2014b) have used the results of the AM3 model to investigate background O₃ at Trinidad Head, California as well as other locations. Meteorological evidence exists to support the observation that conditions representative of US background are routinely encountered at the low-elevation monitoring site at Trinidad Head, California. McDonald-Buller et al. (2011) concluded that conditions representative of background O₃ are routinely encountered at Trinidad Head. The site regularly observes measurements under US background conditions for daytime observations (i.e., mid morning to late afternoon). Long-range transport outside of North America and natural processes, such as stratospheric enhancement, contribute to O₃ concentrations measured at this site.

At Trinidad Head, there is continuous surface O₃ monitoring as well as an ozonesonde station. The ozonesonde observing protocol is to make weekly soundings. However, during the

In evaluating the contribution of NAB O₃ to the observed O₃ at Lassen Volcanic National Park and Sacramento a comparison with both ozonesonde locations is shown (Figs. 12 and 13).

The comparison of the surface MDA8, adjusted NAB, and adjusted O3S O₃ at Trinidad Head with ozonesonde data at 0.5 km (a level in the boundary layer) and 1.0 km (near the top of the boundary layer) indicates that the measured MDA8 surface value is representative of a mixed boundary layer with the surface values closely matching the 0.5 km ozonesonde value and slightly below the 1.0 km value. The adjusted NAB O₃ is the major portion of the observed value. The ozonesonde value at 1.0 km is representative of background air reaching the coast without significant surface O₃ loss. NAB O₃ is always \leq the 1.0 km value and is often near the 0.5 km or surface value. After the bias adjustment, NAB O₃ is very consistent with expected values for the measured O₃ value from the ozonesondes that mostly represent air parcels that have been over the Pacific Ocean for at least several days. The NAB O₃ time series for Trinidad Head for April and May showed that MDA8 O₃ values ranged generally between 30-45 ppb. It is noteworthy that though NAB is the primary contributor to measured O₃ at Trinidad Head, O3S is not the major contributor to NAB. This is consistent with the Lin et al. (2012) results that did not find stratospheric intrusions to significantly influence surface values at Trinidad Head in the spring of 2010.

**Comparing Adjusted AM3 MDA8 Stratospheric O₃,
NAB O₃, and Observed O₃ with sonde 1.5 km
Lassen Volcanic National Park, California**

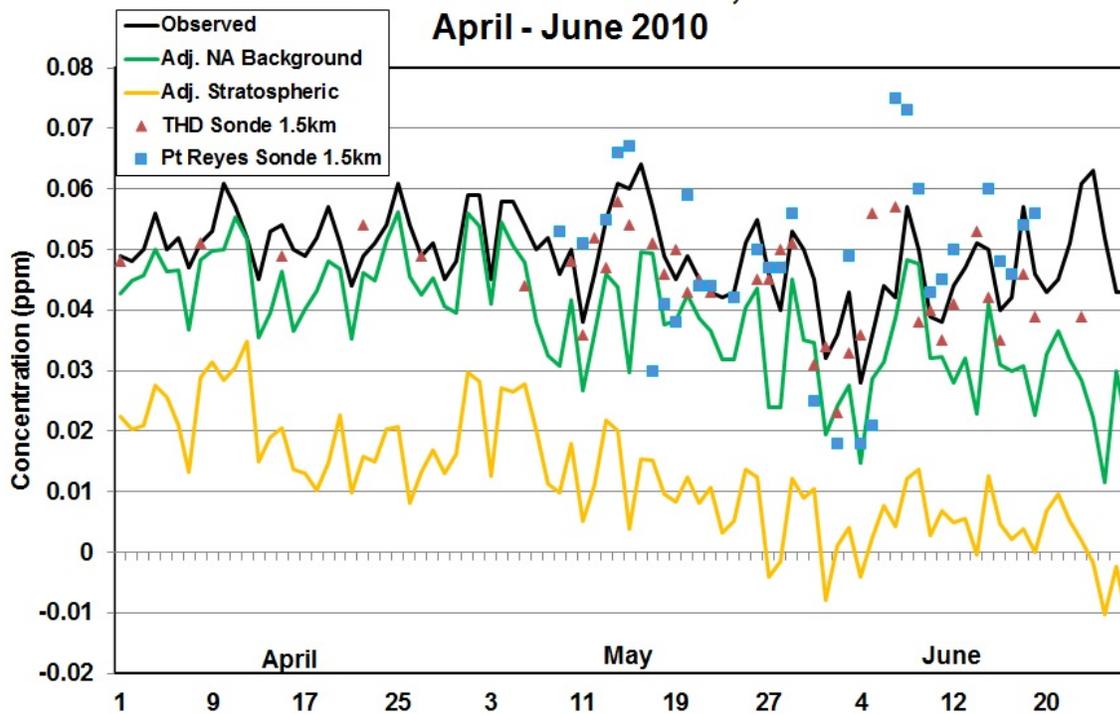


Fig. 12. Comparison of the adjusted AM3 model MDA8 stratospheric O₃ and NAB O₃ and the observed MDA8 O₃ at Lassen Volcanic National Park, CA with ozonesonde measured O₃ 1.5 km at Trinidad Head, CA and Pt. Reyes, CA. Ozonesonde data are 100 m averages centered at the designated altitude. (Source: Lefohn et al., 2014b).

**Comparing Adjusted AM3 MDA8 Stratospheric O₃,
NAB O₃, Observed O₃ and Sonde O₃
Sacramento County, California**

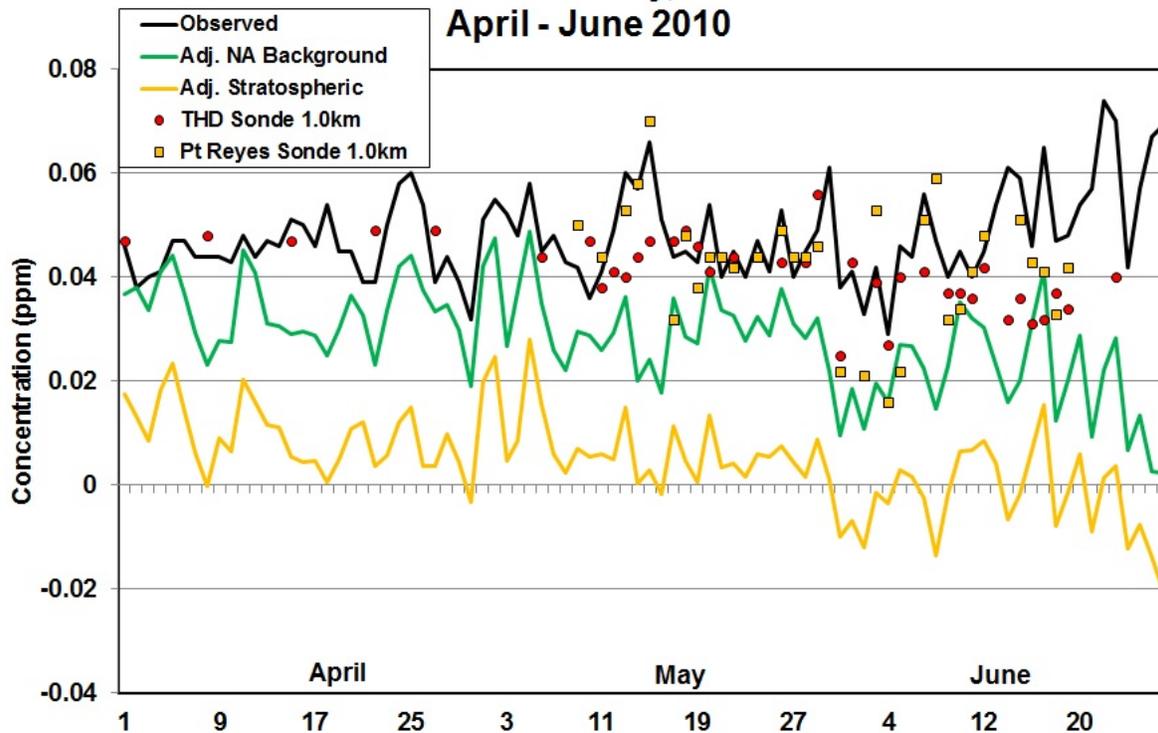


Fig. 13. Comparison of the adjusted AM3 model MDA8 stratospheric O₃ and NAB O₃ and the observed MDA8 O₃ at Sacramento County, CA with ozonesonde measured O₃ 1.0 km at Trinidad Head, CA and Pt. Reyes, CA. Ozonesonde data are 100 m averages centered at the designated altitude. (Source: Lefohn et al., 2014b).

Lassen Volcanic National Park, CA is a higher elevation site (~1.5 km) inland from Trinidad Head. During the spring with prevailing westerly flow it is expected that the Lassen location intercepts air flowing inland from the Pacific. This implies that MDA8 surface O₃ at Lassen should be consistent with O₃ measured entering the west coast of the U.S. at the appropriate altitude (~1.5 km). Comparison of the ozonesonde data with Lassen measured and modeled MDA8 O₃ (Fig. 12) is very consistent with this picture. It shows that the adjusted model NAB O₃ for Lassen generally is well represented by the 1.5 km Trinidad Head ozonesonde value. On the other hand, the Pt. Reyes ozonesonde data show several cases with higher 1.5 km

values that are not reflected in the adjusted NAB or O3S O₃. The higher Pt. Reyes values were on days with noted stratospheric influence at Pt. Reyes but not at Trinidad Head. The large adjusted model NAB contribution to the observed values at Lassen is very consistent with the expectation from the ozonesondes. As at Trinidad Head, the NAB at Lassen is not driven by exceptional contributions from O3S, although adjusted O3S is nearly half of the NAB O₃ on several occasions. The NAB O₃ time series for Lassen for April and May showed that MDA8 O₃ values ranged generally between 20-55 ppb.

At Sacramento County, CA the degree to which the coastal ozonesonde values at a particular altitude are related to the surface MDA8 O₃ and adjusted NAB O₃ is not likely to be as large as at the less locally influenced site at Lassen. Comparing the Sacramento data with the 1.0 km ozonesonde data at the two sites does suggest that the model adjusted NAB O₃ does not overestimate the contribution to observed MDA8 O₃ (Fig. 13). The NAB O₃ time series for Sacramento County for April and May showed that MDA8 O₃ values ranged generally between 20-45 ppb.

Jefferson County, Colorado located in the Denver metropolitan area shows a large contribution from NAB O₃ to the measured MDA8 value (Fig. 14). Though there are only limited ozonesonde data from the Boulder, CO location, they are consistent with a rather significant contribution of NAB O₃ as shown by the model. The ozonesonde data from near the surface to well above the surface are near the adjusted NAB O₃ value. The NAB O₃ time series for Jefferson County for April to June showed that MDA8 O₃ values ranged generally between

**Comparing Adjusted AM3 MDA8 Stratospheric O₃,
NAB O₃, Observed O₃ and Boulder Sondes
Jefferson County, Colorado
April - June 2010**

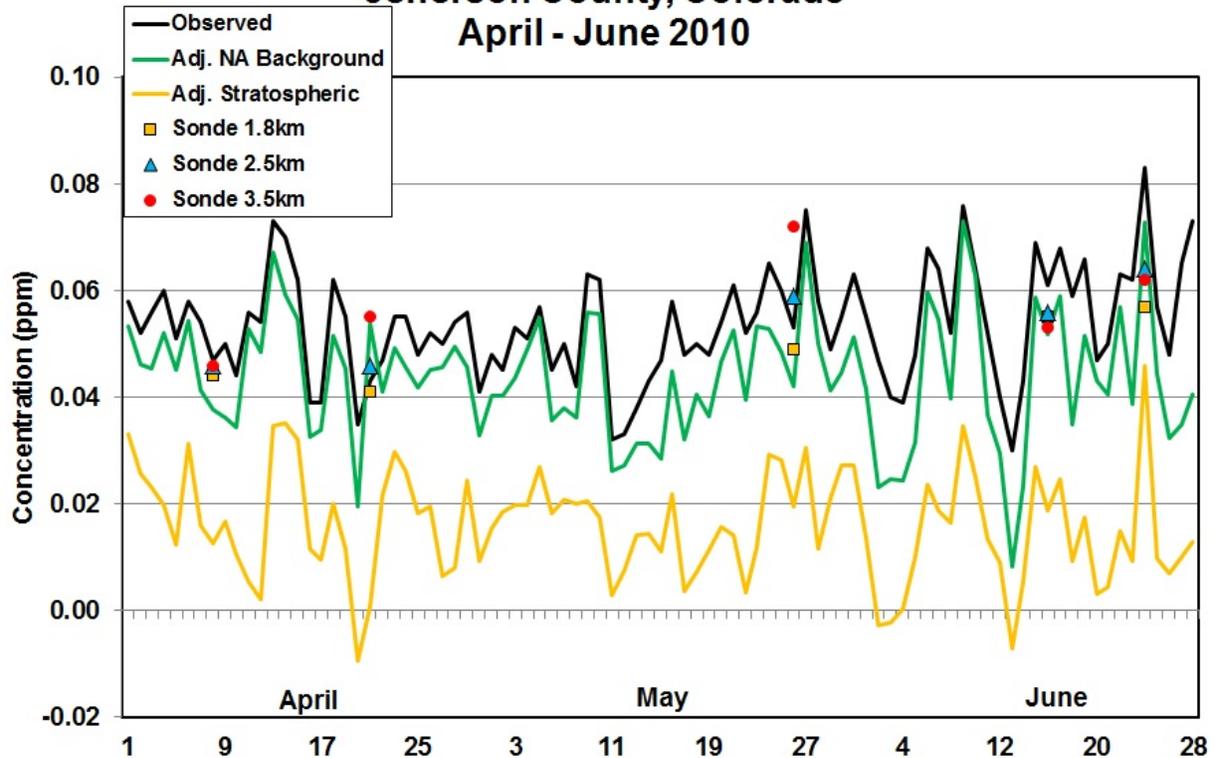


Fig. 14. Comparison of the AM3 model MDA8 stratospheric O₃ and NAB O₃ and the observed MDA8 O₃ at Jefferson County, CO with ozonesonde measured O₃ 1.8 km (near surface). 2.5 km, and 3.5 km at nearby Boulder, CO. Ozonesonde data are 100 m averages centered at the designated altitude. (Source: Lefohn et al., 2014b).

30-70 ppb. The Pinedale, Wyoming site (Fig. 15) does not have a nearby ozonesonde location, but like Jefferson County has a major portion of measured MDA8 O₃ attributed to NAB O₃. On several of the days at Jefferson County and Pinedale, the model shows adjusted O3S to be an important contributor to the adjusted NAB O₃. The NAB O₃ time series for Pinedale for April to June showed that MDA8 O₃ values ranged generally between 30-65 ppb. At Pinedale for the April 9 event with measured MDA8 >70 ppb, the adjusted O3S is ~50 ppb. The LAGRANTO trajectory model (Lefohn et al., 2014) also found that this event showed significant stratospheric influence (Fig. 16). However, several other days with measured MDA8 O₃ over >60 ppb had

**Comparing Adjusted AM3 MDA8 Stratospheric O₃,
NAB O₃, and Observed O₃
Pinedale, Wyoming
April - June 2010**

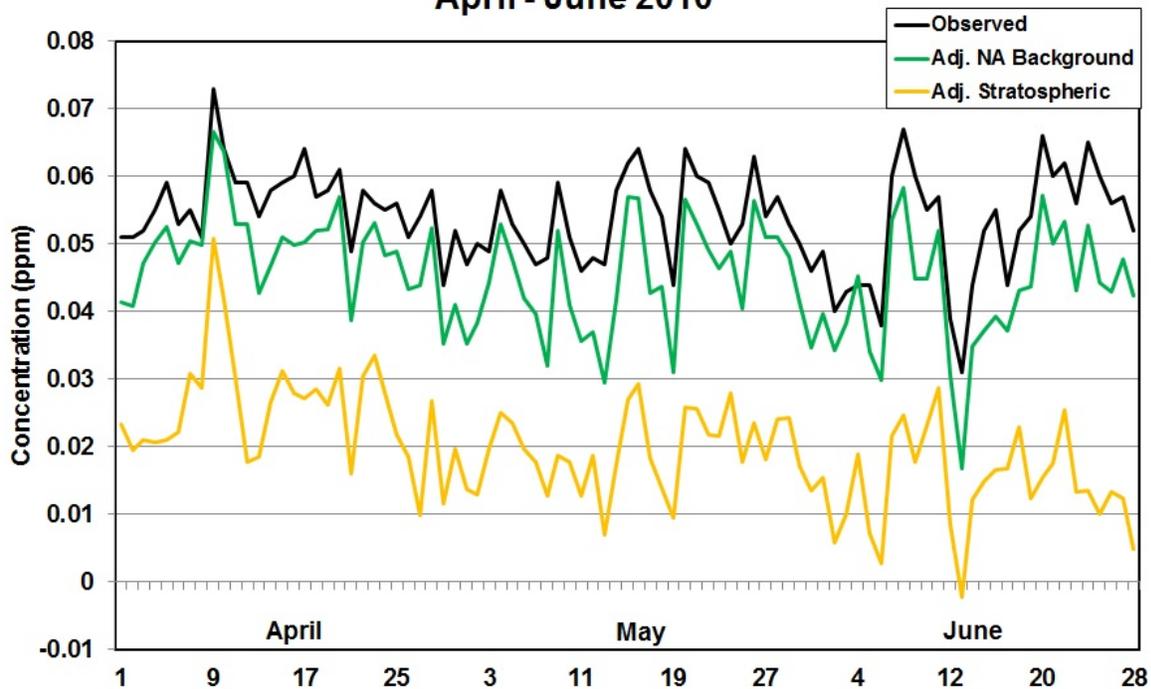


Fig. 15. Comparison of the AM3 model MDA8 stratospheric O₃ and NAB O₃ and the observed MDA8 O₃ at Pinedale, WY. (Source: Lefohn et al., 2014b).

much less of an O₃S contribution though NAB was a major portion of the observed value. This was the case at Jefferson County as well (Fig. 14).

Observed O₃, Unadjusted O₃S, and Daily Total STT-S Counts Pinedale, Wyoming April 6 - 12, 2010

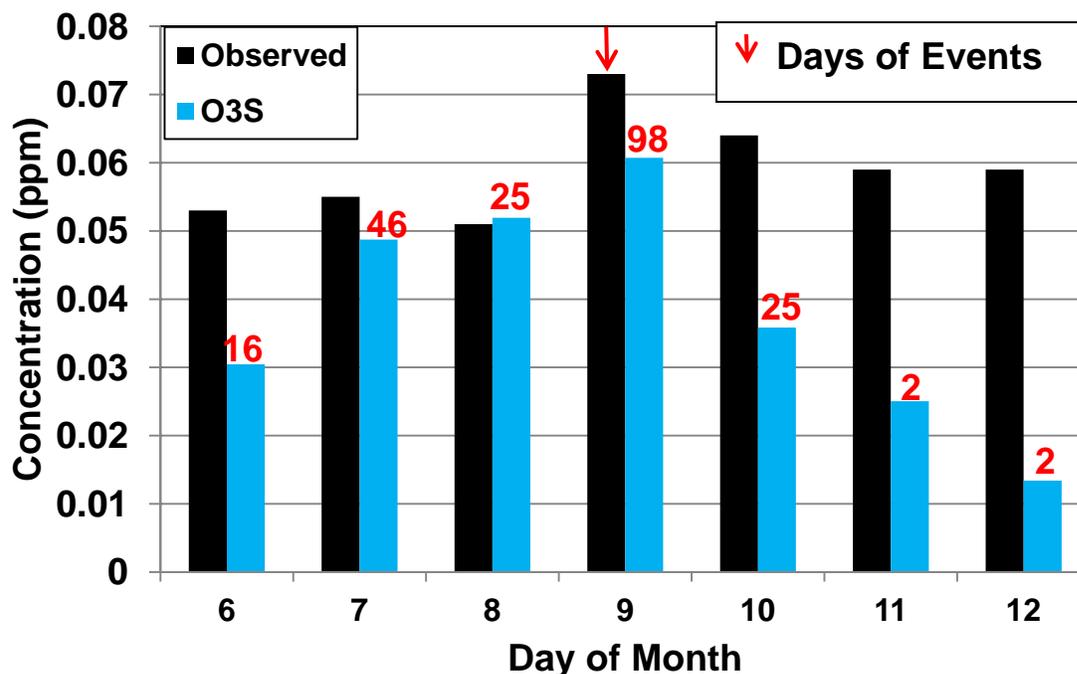


Fig. 16. Event on April 9 at Pinedale, WY, where both the AM3 model and the LAGRANTO trajectory analysis (STT-S counts) show a significant contribution from stratospheric O₃ to NAB O₃. (Note: The O3S O₃ is not adjusted for bias and thus, shows an overly large contribution from the stratosphere. From Fig. 15, the adjusted O3S value for April 9 is ~0.05 ppm). (Source: Lefohn et al., 2014b).

Using results from the GEOS-Chem/CAMx model (Lefohn et al., 2014a) and the ozonesonde data from several sites to evaluate the AM3 model performance (Lefohn et al., 2014b) in apportioning background O₃ (i.e., EIB O₃ and NAB O₃) to the total measured MDA8 O₃ at several sites produces realistic values. For estimating realistic values of EIB O₃ and NAB O₃, adjustments were required as described in Lefohn et al. (2014a, b) to account for likely under (GEOS-Chem/CAMx) and overestimates (AM3). Overall our evaluation shows that the elevated values of NAB O₃ are a consistent feature of spring measured O₃ in the western US. While

several cases could be identified with a relatively large contribution of O₃S to NAB O₃ when measured MDA8 was >60 ppb, in the majority of cases no exceptional cause for high measured values was identified. Elevated NAB O₃ should be seen as the primary contributor to both the high average spring and early summer O₃ in the western US but also to MDA8 amounts >60 ppb.

In the analysis of background O₃ we have described in this section, we found that background O₃ was generally in the range of 30-70 ppb for the high-elevation sites and 30-45 ppb for the Trinidad Head, low-elevation site. A similar range was observed for other low-elevation sites. As we discussed earlier, the lung function and epidemiological risk estimates for attaining the 75, 70, 65 and 60 ppb standards indicate that a large percentage of the risks are associated with 8-hour average ambient concentrations in the 25-55 ppb range, which is the range of concentrations associated with background O₃.

References

- 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.
- Fiore, A. M., Jacob, D.J., Bey, I., Yantosca, R.M., Field, B.D., Fusco, A.C., Wilkinson, J.G., 2002. Background ozone over the United States in summer: Origin, trend, and contribution to pollution episodes. *Journal of Geophysical Research* 107(D15), 4275, doi:10.1029/2001JD000982.
- Fiore, A., Jacob, D.J., Liu, H., Yantosca, R.M., Fairlie, T.D., Li, Q., 2003. Variability in surface ozone background over the United States: Implications for air quality policy. *Journal of Geophysical Research* 108 (D24), 4787, doi:10.1029/2003JD003855.
- Fiore, A.M., Oberman, J.T., Lin, M., Zhang, L., Clifton, O.E., Jacob, D.J., Naik, V., Horowitz, L.W., Pinto, J.P. 2014. Estimating North American background ozone in U.S. surface air with two independent global models: Variability, uncertainties, and recommendations. (Submitted).
- Lapina, K., Henze, D.K., Milford, J.B., Huang, M., Lin, M., Fiore, A.M., Carmichael, Pfister, G.G., Bowman, K., 2014. Assessment of source contributions to seasonal vegetative

exposure to ozone in the U.S. *Journal of Geophysical Research*. 119:324-340.
DOI: 10.1002/2013JD020905.

Lefohn A. S., Shadwick D.S. and Ziman S.D., 1998. The Difficult Challenge of Attaining EPA's New Ozone Standard. *Environmental Science & Technology*. 32(11):276A-282A.

Lefohn, A.S., Shadwick, D., Oltmans, S.J., 2008. Characterizing long-term changes in surface ozone levels in the United States (1980-2005). *Atmospheric Environment* 42, 8252-8262.
<http://dx.doi.org/10.1016/j.atmosenv.2008.07.060>.

Lefohn, A.S., Shadwick, D., Oltmans, S.J., 2010. Characterizing changes in the distribution of surface ozone levels in metropolitan and rural areas in the United States for 1980-2008 and 1994-2008. *Atmospheric Environment* 44, 5199-5210.
<http://dx.doi.org/10.1016/j.atmosenv.2010.08.049>.

Lefohn, A.S., Oltmans, S.J., 2012. Background Ozone and Its Importance in Relation to Health Risk and Exposure Assessment for Ozone Assessment Document. Submitted to the Docket ID No. EPA-HQ-OAR-2008-0699. September 17, 2012.

Lefohn, A.S., Emery, C., Shadwick, D., Wernli, H., Jung, J., Oltmans, S.J., 2014a. Estimates of Background Surface Ozone Concentrations in the United States Based on Model-Derived Source Apportionment. *Atmospheric Environment*,
<http://dx.doi.org/10.1016/j.atmosenv.2013.11.033>. 84: 275-288.

Lefohn, A.S., Shadwick, D., Oltmans, S.J., Wernli, H., Lin, M. 2014b. Characterizing the relative importance of stratospheric and background to observed ozone in the US with the GFDL AM3 Chemistry-Climate Model combined with the LAGRANTO trajectory-based approach. Manuscript in preparation.

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., 2012. 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.

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, doi:10.1021/es2022918, 9484-9497.

Oltmans, S.J., Lefohn, A.S., Harris, J.M., Galbally, I., Scheel, H.E., Bodeker, G., Brunke, E., Claude, H., Tarasick, D., Johnson, B.J., Simmonds, P., Shadwick, D., Anlauf, K., Hayden, K., Schmidlin, F., Fujimoto, T., Akagi, K., Meyer, C., Nichol, S., Davies, J., Redondas, A., Cuevas, E., 2006. Long-term changes in tropospheric ozone. *Atmospheric Environment* 40, 3156e3173. <http://dx.doi.org/10.1016/j.atmosenv.2006.01.029>.

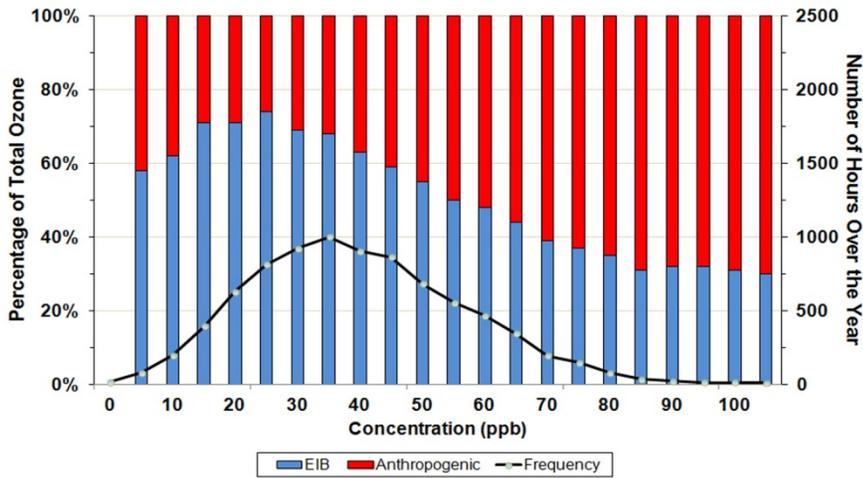
- Oltmans, S.J., Lefohn, A.S., Harris, J.M., Shadwick, D.S., 2008. Background ozone levels of air entering the west coast of the U.S. and assessment of longer-term changes. *Atmospheric Environment* 42, 6020-6038. <http://dx.doi.org/10.1016/j.atmosenv.2008.03.034>.
- Oltmans, S.J., Lefohn, A.S., Shadwick, D., Harris, J.M., Scheel, H.-E., Galbally, I., Tarasick, D.A., Johnson, B.J., Brunke, E., Claude, H., Zeng, G., Nichol, S., Schmidlin, F., Redondas, A., Cuevas, E., Nakano, T., Kawasato, T., 2013. Recent Tropospheric Ozone Changes – A Pattern Dominated by Slow or No Growth. *Atmospheric Environment*. doi:10.1016/j.atmosenv.2012.10.057. 67: 331-351.
- US Environmental Protection Agency, US EPA, 1996. Review of National Ambient Air Quality Standards for Ozone - Assessment of Scientific and Technical Information, OAQPS Staff Paper. EPA-452/R-96-007, Office of Air Quality Planning and Standards, Research Triangle Park, NC. June 1996. US Environmental Protection Agency.
- US Environmental Protection Agency, US EPA, 2006. Air quality criteria for ozone and related photochemical oxidants. Report Nos. EPA/600/R-05/004af, Office of Research and Development, Research Triangle Park, NC. February 2006. US Environmental Protection Agency.
- US Environmental Protection Agency, US EPA, 2013. Integrated Science Assessment for Ozone and Related Photochemical Oxidants. EPA/600/R-10/076F. Research Triangle Park, NC: Office of Research and Development. February.
- US Environmental Protection Agency, US EPA, 2014a. Health Risk and Exposure Assessment for Ozone. Second external review draft. EPA/452/P-14-004a. Research Triangle Park, NC: Office of Air Quality Planning and Standards. January.
- US Environmental Protection Agency, US EPA, 2014b. Policy Assessment for the Review of the Ozone National Ambient Air Quality Standards. Second external review draft. EPA/452/P-14-002. Research Triangle Park, NC: Office of Air Quality Planning and Standards. January.
- 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 $1/2^\circ \times 2/3^\circ$ horizontal resolution over North America. *Atmospheric Environment* 45, 6769-6776.
- Zhang, L., Jacob, D.J., Yue, X., Downey, N.V., Wood, D.A., Blewitt, D., 2013. Sources contributing to background surface ozone in the US Intermountain West. *Atmos. Chem. Phys. Discuss.*, 13, 25871–25909, 2013 www.atmos-chem-phys-discuss.net/13/25871/2013/ doi:10.5194/acpd-13-25871-2013

Appendix

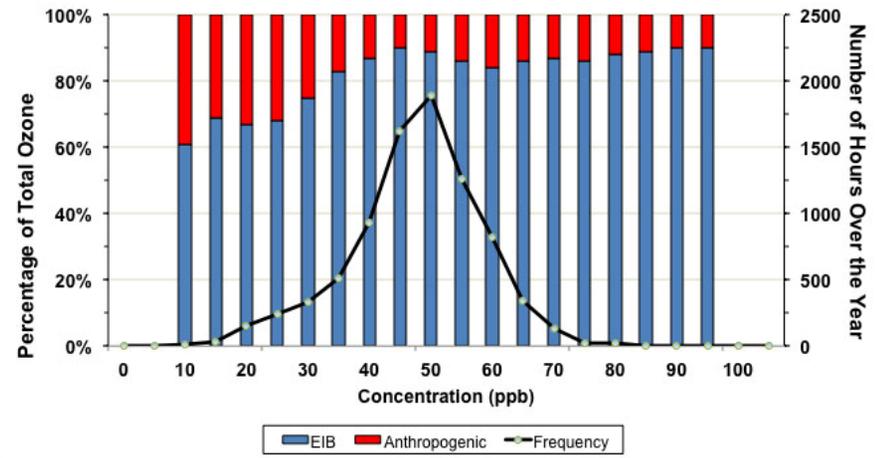
Anthropogenic and Emissions-Influenced Background (EIB) Contributions to Total Ozone Concentrations for 2006

Source: Lefohn et al. (2014a)

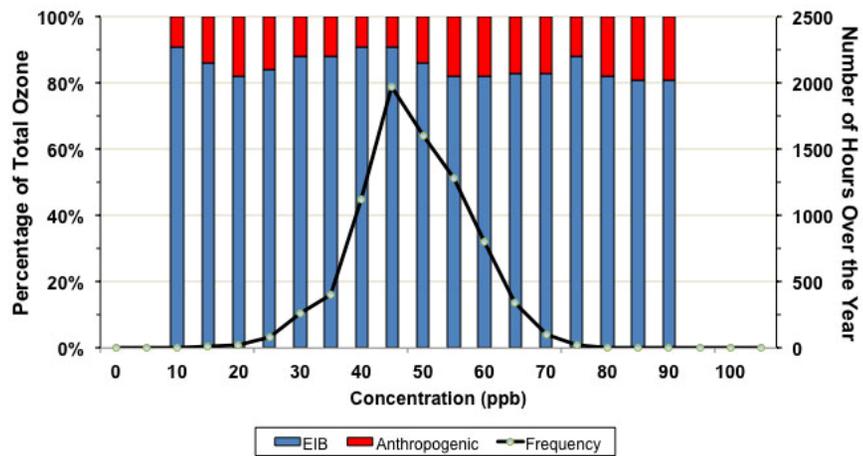
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Data for the Year 2006**



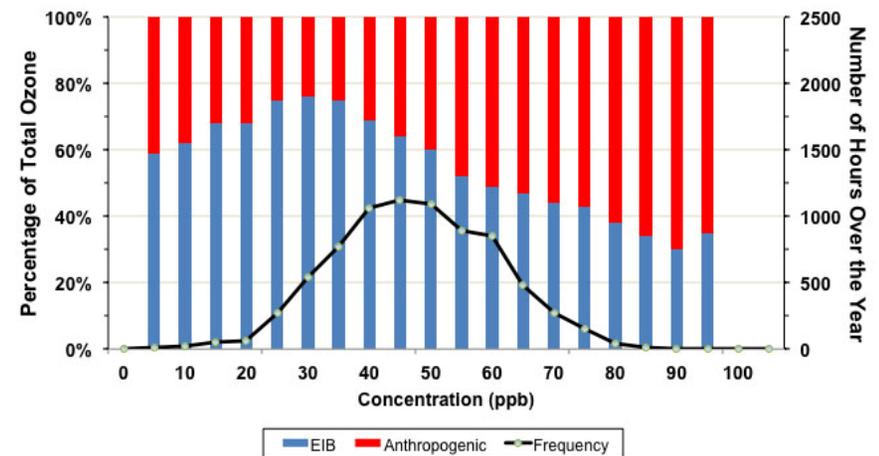
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Data for the Year 2006**



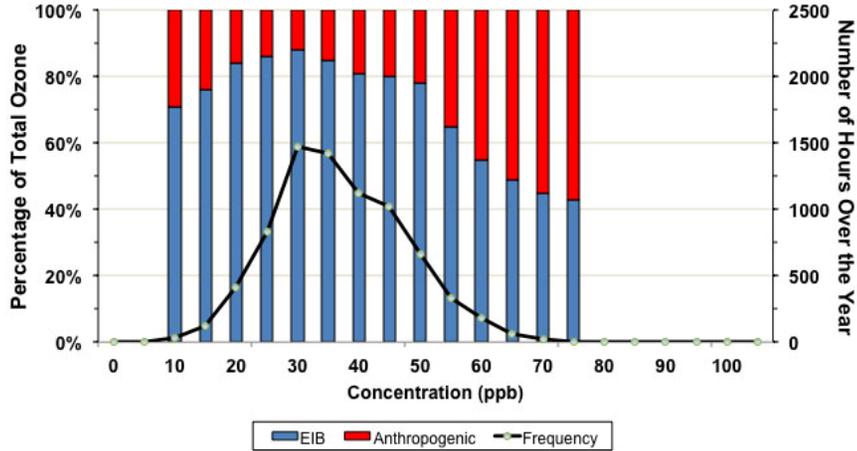
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Data for the Year 2006**



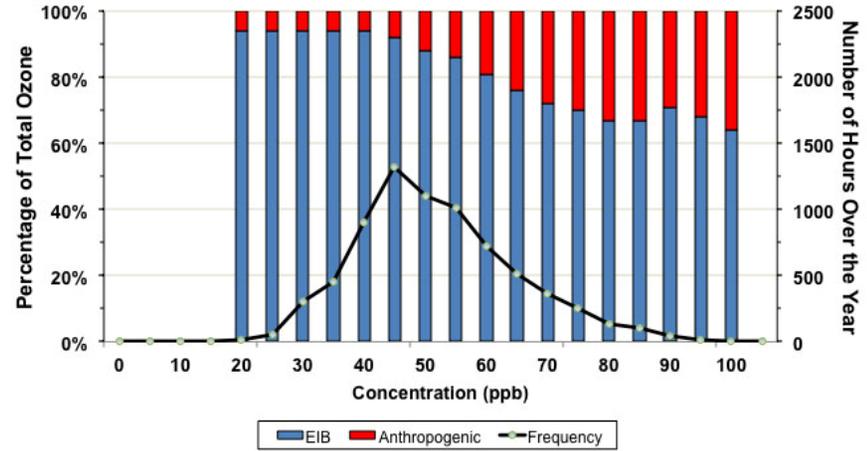
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Shenandoah NP, VA
Data for the Year 2006**



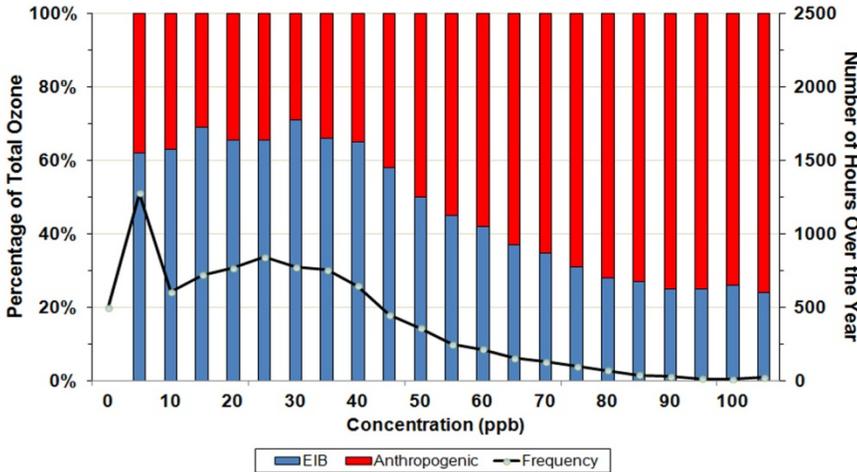
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Data for the Year 2006**



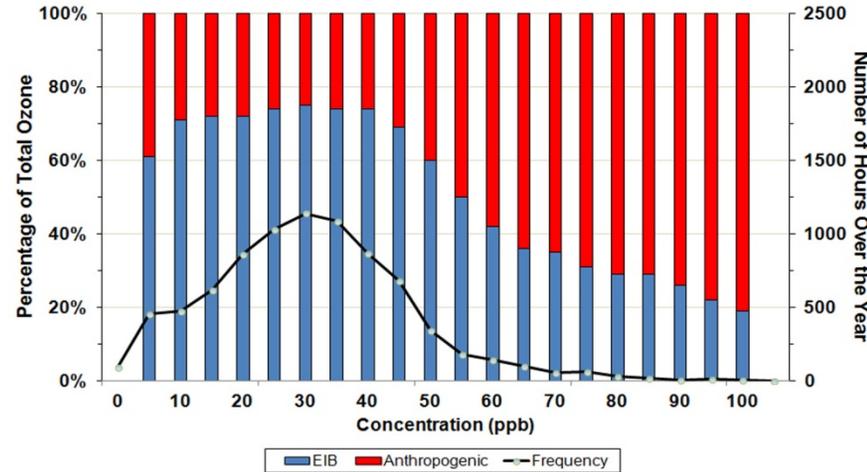
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Data for the Year 2006**

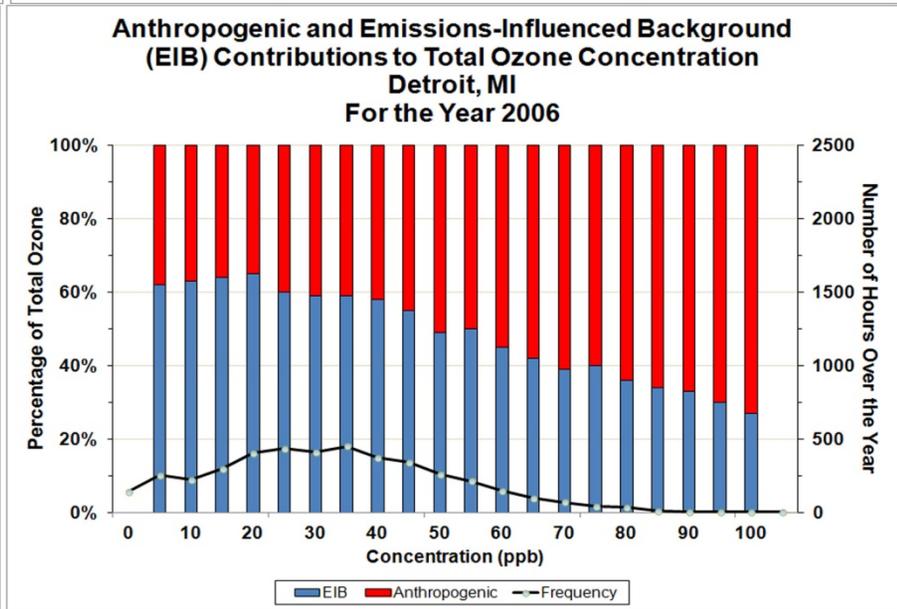
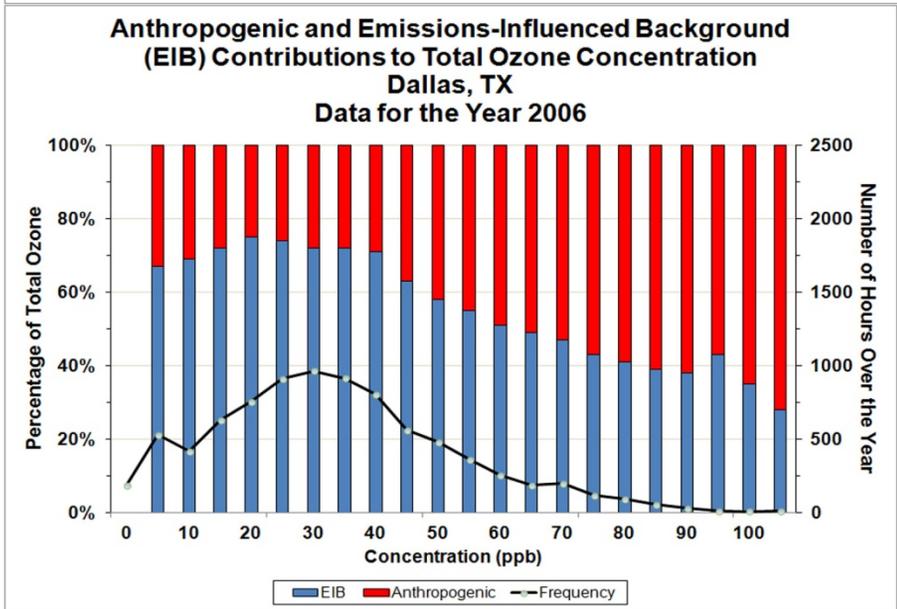
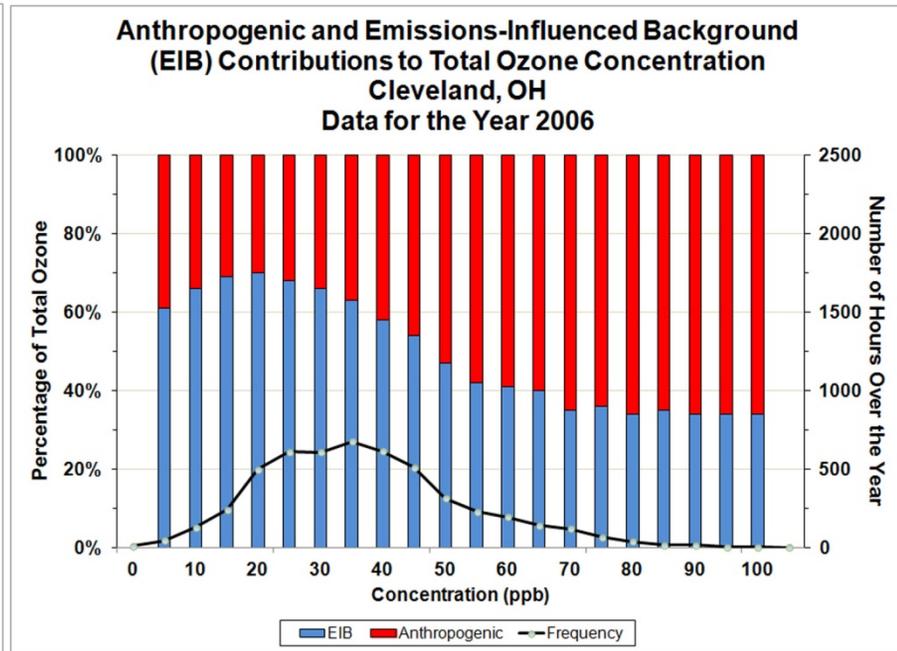
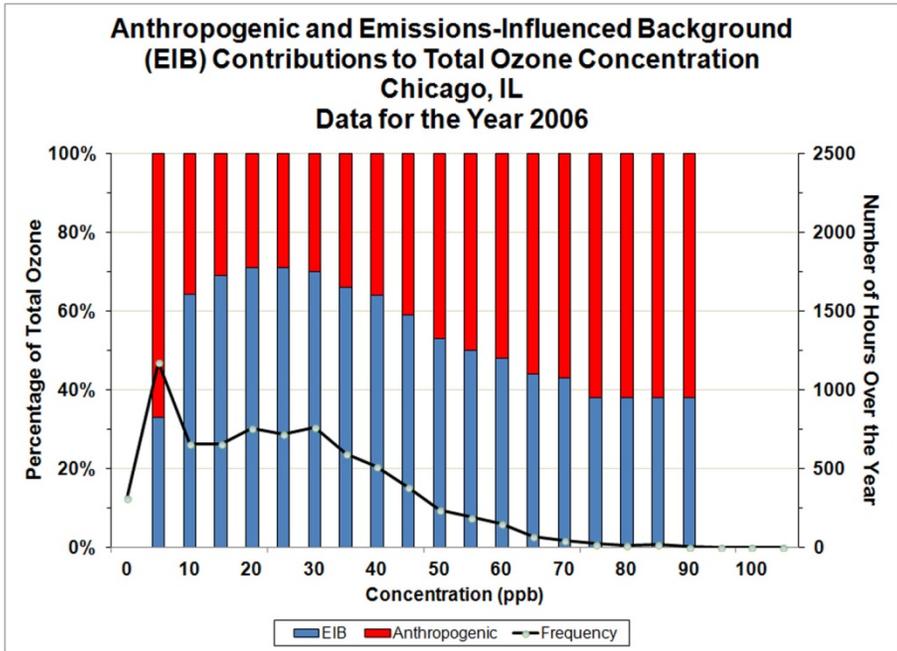


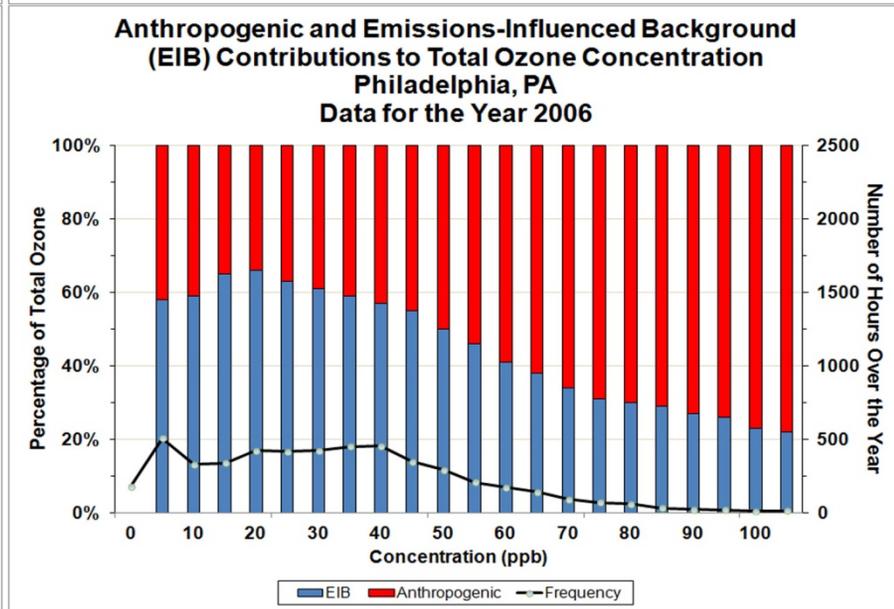
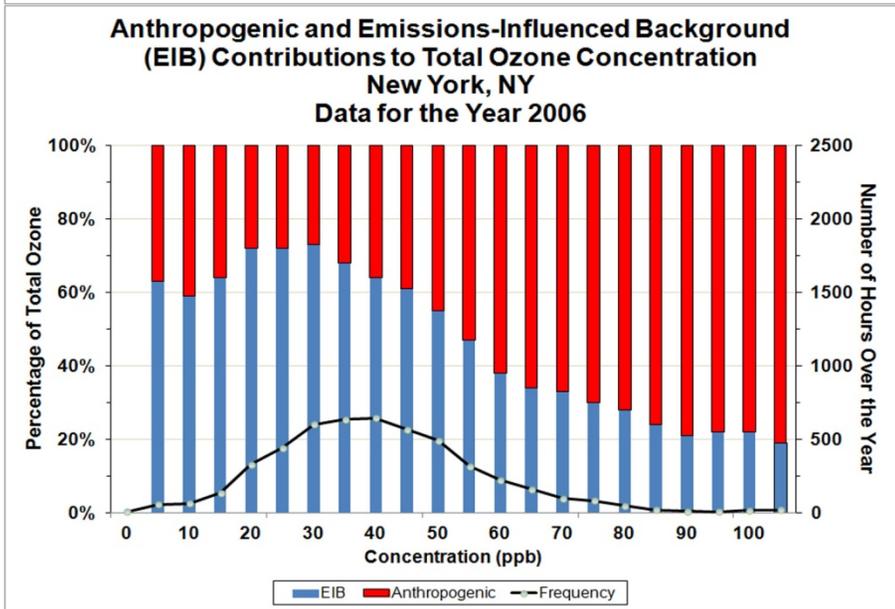
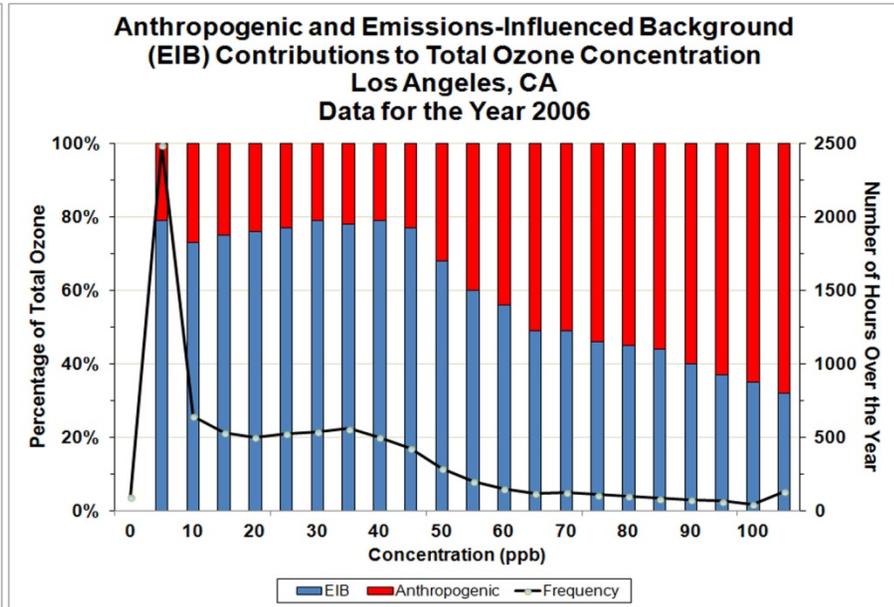
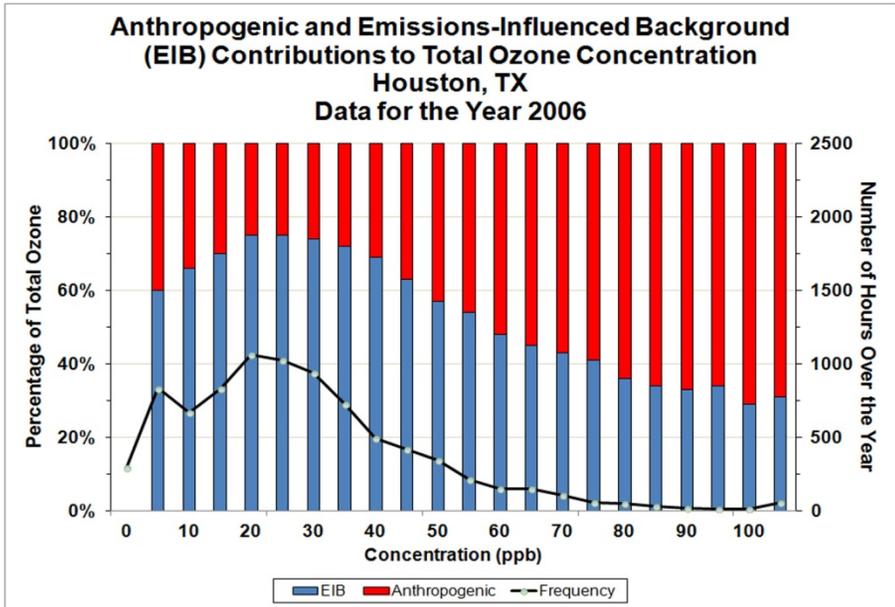
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Baltimore, MD
Data for the Year 2006**

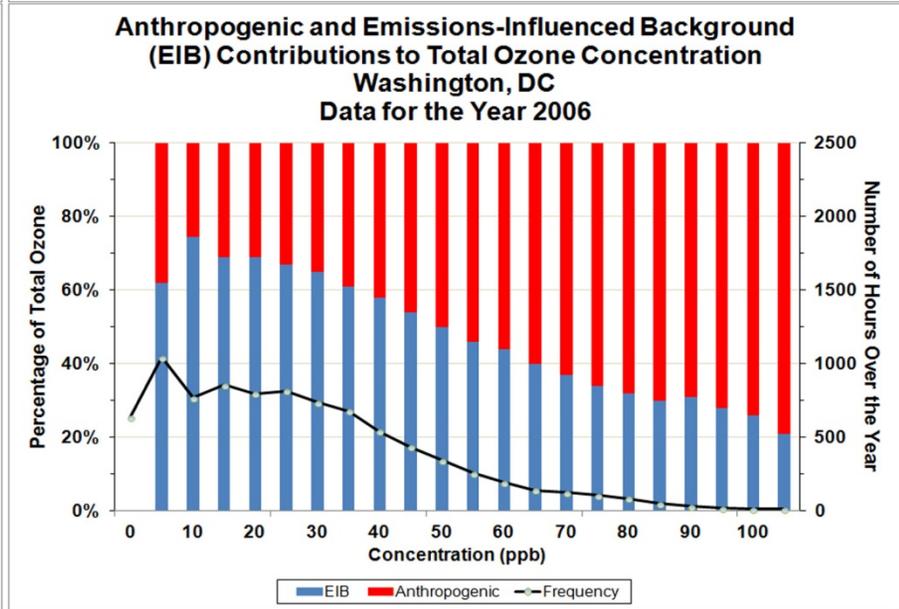
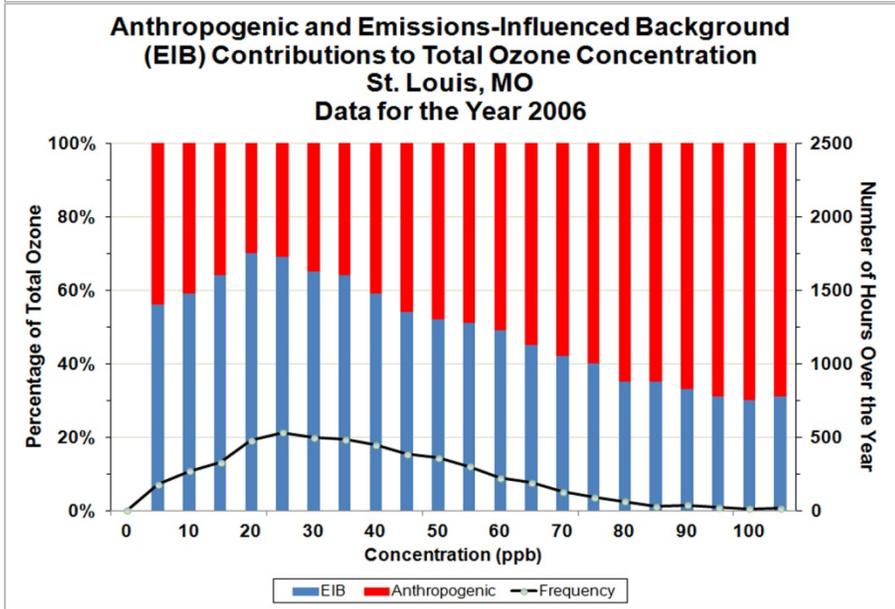
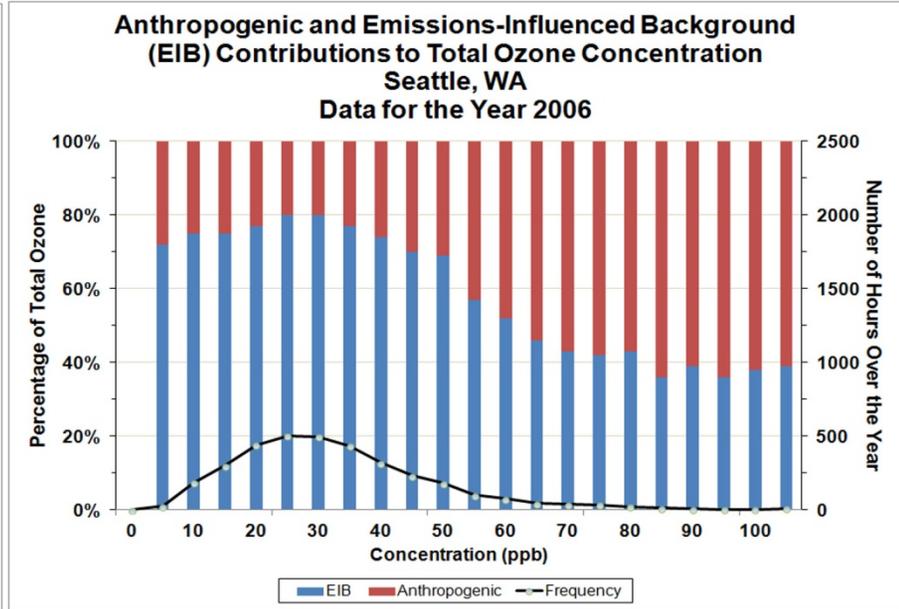
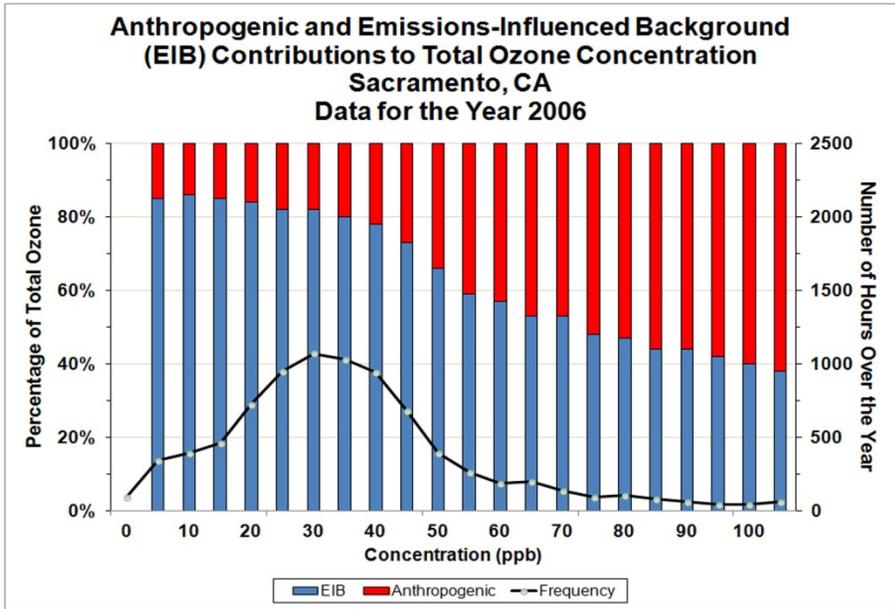


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Boston, MA
Data for the Year 2006**









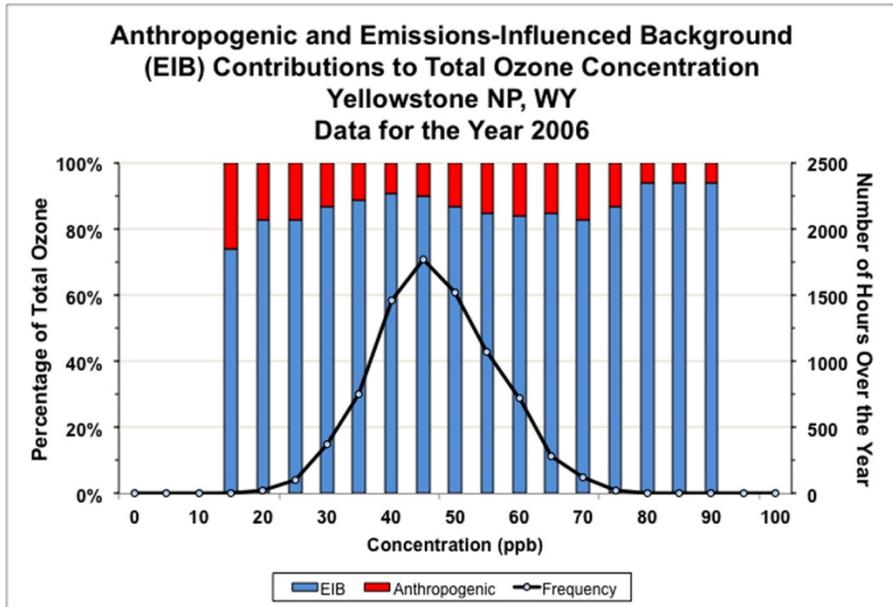
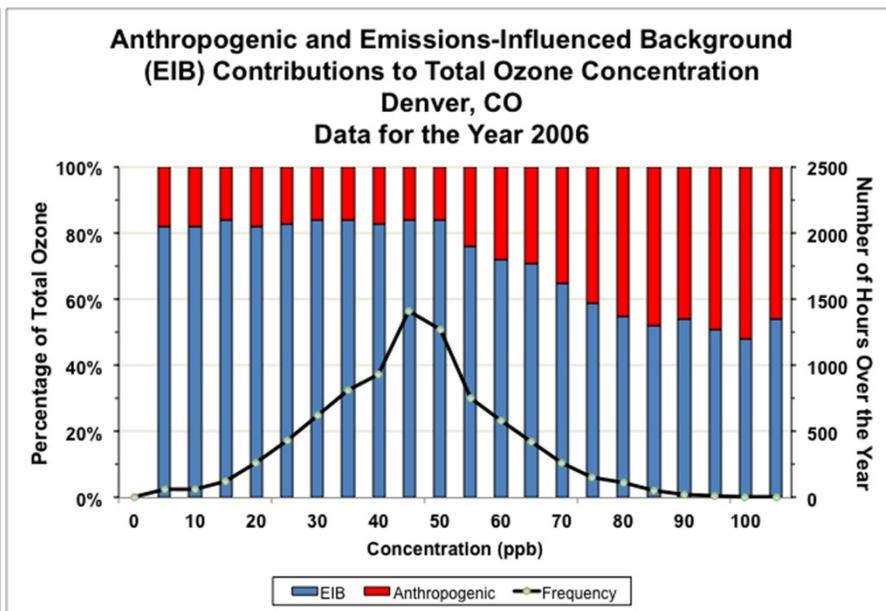
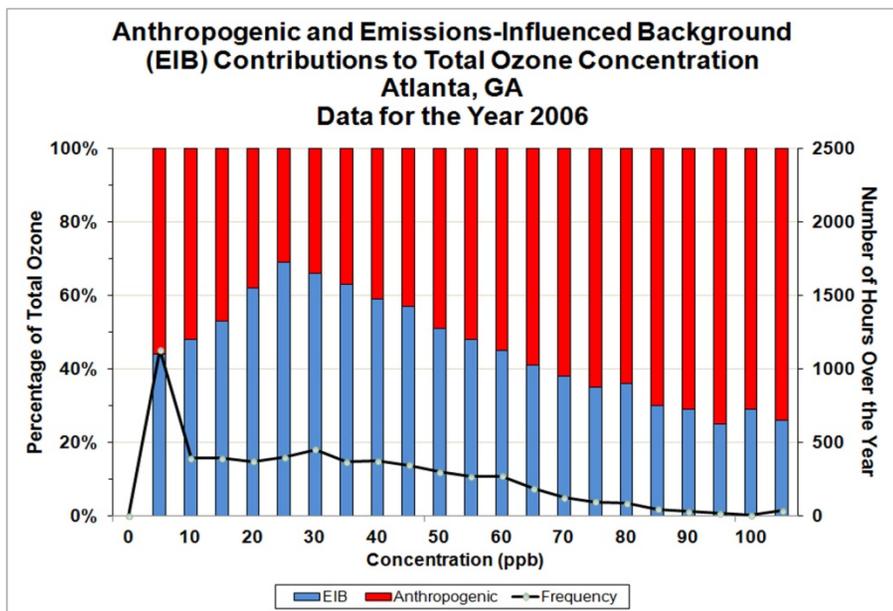


Fig. S1. Binned (5 ppb) frequency distribution of observed hourly total O₃ (black curve; right axis) and average relative binned contributions of hourly maximum EIB and anthropogenic O₃ (bars; left axis) for all other sites analyzed. (Source: Lefohn et al. (2014a).