

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

For Consideration by CASAC

Allen S. Lefohn, A.S.L. & Associates
302 North Last Chance Gulch, Suite 410, Helena, Montana 59601
asl-associates.com

Samuel J. Oltmans
1619 Pine Street, Boulder, CO 80302

August 31, 2012

ES. Executive Summary

ES-1. The Importance of Background Ozone for Risk Assessment

- The uncertainty associated with the estimates of background O₃ has an important role in developing confidence in the risk assessment results described in Chapter 6 and Chapter 7.
- The level of estimated U.S. background is also important when assessing the reliability of the estimates of the Lowest Measured Level (LML).
- Using the Heat Map (Table 7-9), 9 of the 12 cities are predicted by EPA to experience approximately 50% or more of O₃-related all-cause mortality based on epidemiological results for daily 8-h maximum O₃ levels between 25 and 50 ppb. Measured background O₃ concentrations are in the range of 25 to 50 ppb and at times, higher.
- For the second draft of the REA, the EPA Agency assumes that estimates of background can be obtained directly from modeling. While acknowledging that the quadratic rollback method has its limitations, ignoring the limitations associated with the Higher-order Decoupled Direct Method (HDDM) methodology may lead to great uncertainty in the estimation of U.S. background. If the model does not contain modules that can adequately address natural processes that contribute to background, such as stratospheric-tropospheric exchange, estimates of U.S. background levels will likely be subject to great uncertainties, which in turn, can affect risk estimates.
- The range of U.S. background level concentrations is important when characterizing health risks based on clinical studies. The REA illustrates the probabilistic exposure-response relationships for FEV₁ decrement $\geq 10\%$. Although the probability of a response below 50 ppb is low, a large fraction of the population is estimated to be exposed to these low O₃ concentrations. These low O₃ concentrations are used in the risk model to predict the number of persons responding. If U.S. background O₃

concentrations are much larger than predicted by the CMAQ model, then the risk predicted for this large fraction of the population may not be able to be reduced if reductions in emissions were to occur.

ES-2. Comparing Model Results with Observational Data

- The EPA notes that currently all chemical transport models and not just the ones evaluated in the ISA, have difficulty in predicting hourly or 8-h daily maximum concentrations.
- When model results are compared with observational data, the models are unable to adequately capture the O₃ concentrations enhancements (i.e., hourly average concentrations ≥ 50 ppb) associated with stratospheric intrusions and biomass burning.
- Estimations of hourly averaged O₃ concentrations of U.S. background, as well as NA background, is a model construct that must be informed by and evaluated based on observational data.
- Similarly, the model-constructed and mathematically smoothed diurnal patterns of U.S. background O₃ concentrations that EPA uses in its risk assessments must also be informed by and evaluated based on observational data.
- One very important set of relevant observational data available describe the hourly average O₃ concentrations collected at Trinidad Head, California. Although the ISA in its third draft, similar to the other two drafts, continues to indicate that recent North American emissions contribute significantly to O₃ measured at coastal sites, such as Trinidad Head, this statement is inaccurate and is not supported in the peer-reviewed published literature. Meteorological evidence exists to support the observation that conditions representative of U.S. background are routinely encountered at the low-elevation monitoring site at Trinidad Head, California. Trinidad Head regularly observes measurements under U.S. 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.
- The frequency of hourly average concentrations ≥ 50 ppb in the springtime, when almost all of the high concentrations occur, is large and varies from year to year during the month of April (e.g., 30 to 187 hours) (see Table E-1 below). The range of maximum hourly average O₃ concentrations for April over the period 2002-2011 is 54 – 65 ppb. For the period 2003-2011, daily maximum 8-h average O₃ concentrations at times exceed 60 ppb during the springtime. The range of 3-year average of the 4th highest maximum 8-h average concentrations is from 50 to 52 ppb.

- The O₃ values at Trinidad Head are indicative of O₃ amounts reaching the California coast from the Pacific.
- When compared to the observational data at Trinidad Head, GEOS-Chem, model predictions for the spring indicate that daily maximum 8-h average O₃ concentrations are consistently underestimated (see Fig. E-1), which suggests that model background levels are estimated to be too low for this season.

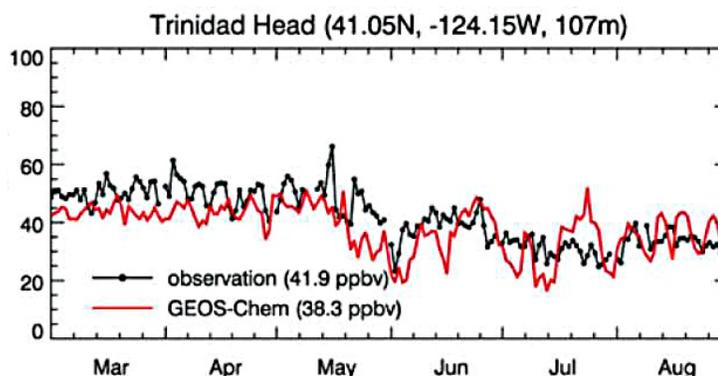


Fig. E-1. Comparison of daily maximum 8-h average O₃ predicted using GEOS-Chem at 0.5° × 0.667° with measurements at Trinidad Head, CA from March to August 2006. Source: US EPA (2012a).

Table E-1. Summary of the number of hourly average concentrations ≥ 0.05 ppm and maximum hourly average value (in parentheses) measured at Trinidad Head, California for the 10-year period, 2002 through 2011. UTC time period.

Year	March	April	May	June	July
2002	No Data	36 (54 ppb)	18 (53 ppb)	0 (41 ppb)	0 (45 ppb)
2003	23 (52 ppb)	187 (64 ppb)	96 (57 ppb)	13 (54 ppb)	0 (41 ppb)
2004	38 (62 ppb)	30 (54 ppb)	3 (50 ppb)	2 (50 ppb)	0 (38 ppb)
2005	58 (55 ppb)	72 (56 ppb)	6 (52 ppb)	0 (46 ppb)	0 (39 ppb)
2006	37 (55 ppb)	56 (60 ppb)	43 (64 ppb)	0 (47 ppb)	0 (35 ppb)
2007	21 (52 ppb)	46 (57 ppb)	30 (55 ppb)	0 (40 ppb)	0 (33 ppb)
2008	26 (53 ppb)	120 (61 ppb)	34 (56 ppb)	3 (52 ppb)	0 (42 ppb)
2009	18 (54 ppb)	73 (65 ppb)	11 (54 ppb)	0 (43 ppb)	0 (35 ppb)
2010	38 (58 ppb)	51 (58 ppb)	6 (52 ppb)	0 (42 ppb)	0 (32 ppb)
2011	18 (54 ppb)	33 (57 ppb)	5 (51 ppb)	0 (48 ppb)	0 (30 ppb)

- The EPA illustrates in the REA the diurnal profiles of seasonally averaged hourly U.S. background floor values estimated for Los Angeles and Sacramento. The springtime values estimated for Los Angeles and Sacramento are less than 30 ppb. These concentrations are much lower than the actual springtime values observed at

- The Crestline site, located in southern California, is heavily influenced by anthropogenic sources. The site experiences some of the highest O₃ exposures in the US. The Crestline site frequently experiences enhanced O₃ concentrations statistically associated with stratospheric events during the spring and fall months. Langford et al. (2011) has presented evidence that stratospheric-tropospheric exchange processes enhanced surface O₃ concentrations at the site during the 43-day study period. Ozone concentrations associated with STT-S events appear to supplement the enhanced O₃ concentrations that are associated with locally generated anthropogenic sources.
- Results from a preliminary modeling analysis by Lefohn, Emery, and co-investigators support the EPA's observation that chemical transport models have difficulty in predicting day-specific concentrations. Hourly observation data at 8 O₃ monitoring sites were compared with model predictions. The authors indicate that hourly average total and background O₃ levels tend to be under predicted during late winter through early summer using the GEOS-Chem/CAMx model. The authors superimposed over the model observation comparison a tabulation of monthly stratospheric-tropospheric transport to the surface (STT-S) counts based on Lagrangian trajectory modeling as described in Lefohn et al. (2011; Lefohn et al., 2012-submitted).
- Preliminary analysis by Lefohn, Emery, and co-investigators indicates that the underestimated O₃ occurred when the stratosphere appeared to contribute the most to enhancing hourly average O₃ concentrations ≥ 50 ppb. The monthly coincidences between enhanced (i.e., ≥ 50 ppb) O₃ concentrations and STT events was statistically significant during this period of time. For Yellowstone NP (WY) during the months of March, April, and May of 2006, the site experienced 1,240 occurrences of hourly average O₃ concentrations ≥ 50 ppb. The analysis showed that the months with the largest underestimated median bias were also the months with the largest statistically significant STT contribution. STT counts in Fig. E-2 below are the total number of STT-S hits during the month. The "*" refers to statistically significant coincidences as described in Lefohn et al. (2011). The preliminary results appear to complement EPA's conclusion that the model used to estimate U.S. background may not be able to adequately capture the full impact of STT enhancements to surface O₃.

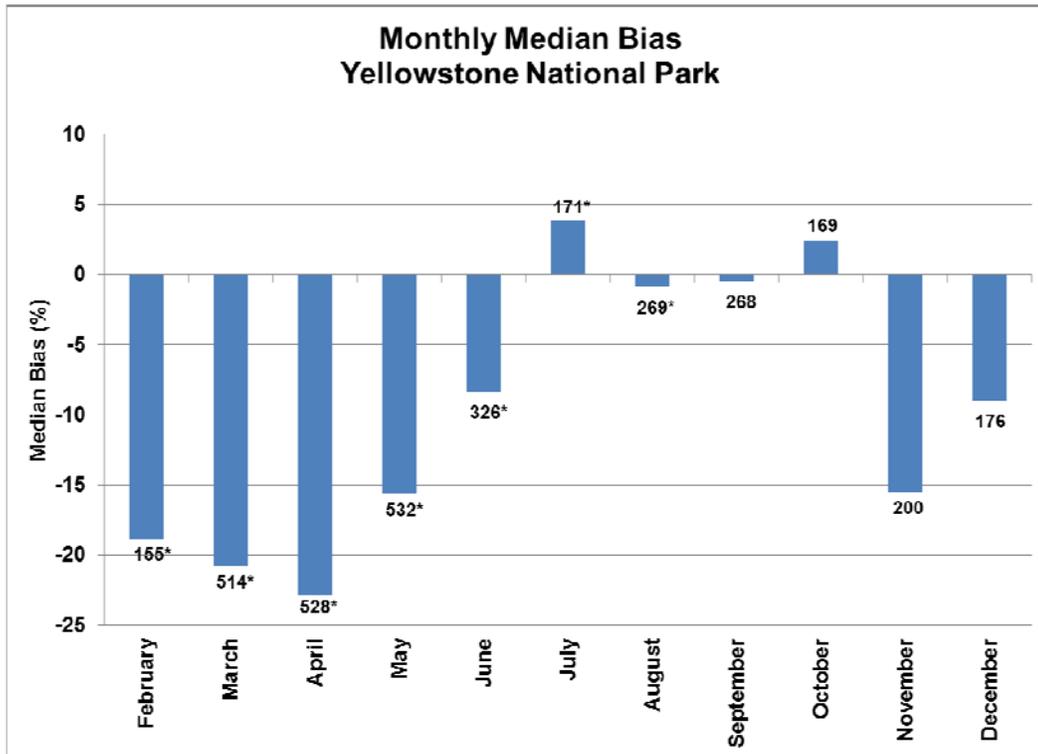


Fig. E-2. Monthly median bias between modeled and observed hourly O₃ at Yellowstone NP for 2006. The total STT-S counts for each month are indicated; a statistically significant month is indicated with an asterisk (*).

ES-3. The Importance of the Stratosphere and Biomass Burning in Affecting Surface Ozone Concentrations

- Evidence suggests that stratosphere-to-troposphere transport to the surface (STT) processes affect surface O₃ concentrations at both high- and low-elevation monitoring sites during the springtime as well as other times during the year more frequently than previous studies may have indicated. STT processes, as well as biomass burning, are likely contributors to background concentrations that can lead to the exceedance of the human-health related O₃ standard. The naturally occurring enhancements associated with STT process have an important effect on estimating hourly average natural background as well as U.S. background O₃ concentrations.
- Results published in the literature indicate that the frequency of STT events that contribute to the variability of natural background is high. At many high- and low-elevation monitoring sites, particularly during the springtime, numerous days occur in which a strong relationship exists between enhanced O₃ concentrations and stratosphere-to-troposphere transport that reaches the surface (STT-S).

- When statistically significant coincidences occur at the lower elevation sites, the daily maximum hourly average concentrations are mostly in the 50-65 ppb range; the daily maximum 8-h average concentrations are usually in the 50-62 ppb range.
- Investigation of specific monitoring sites during periods when they were least affected by days with lightning, long-range transport from Asia, wildfires, or anthropogenic perturbations show that the high-elevation sites experience a greater percentage of days with concentrations ≥ 50 ppb and ≥ 60 ppb on days when STT-S >0 during the springtime.
- Besides STT events, Eurasian emissions associated with biomass burning and their easterly transport contribute to O₃ concentrations at west coast O₃ monitoring sites under U.S. background conditions, as well as inland O₃ monitoring sites in Montana, Wyoming, and North Dakota. Published information indicates that biomass burning events in Eurasia are not infrequent.

ES-4. Applying Recent Advancements in Knowledge to Improve Background Ozone Estimates

- Estimating background, as well as U.S. background O₃ concentrations correctly, is extremely important for assessing risk. The use of diurnal averaging techniques to synthesize hourly concentrations has the potential for estimating extremely low values of background that are not realistic.
- At this time, based upon our evaluation of the science, as well as EPA's conclusions in the ISA, existing models have difficulty in adequately estimating hourly average O₃ concentrations.
- Recent advancements in science have begun to quantify the importance of stratospheric processes in affecting background surface O₃ concentrations.
- It is possible to better characterize background O₃ than the methodology utilized by the EPA for assessing risk.

1. Background Ozone and Risk are Closely Linked Together

In the first draft of the REA (US EPA, 2012a), the EPA implies in estimating the risk from exposure to O₃ it is not necessary to take into consideration U.S. background concentrations beyond application of the quadratic rollback method. For the second draft of the REA, the EPA Agency assumes that estimates of background can be obtained directly from the modeling. This report discusses the reasons why ignoring background is not warranted in either the first or second draft of the REA.

The US EPA notes in the REA (US EPA, 2012a) that the quadratic rollback method has difficulty in adequately representing the complexities associated with emission reductions. The rollback methodology reduces O₃ concentrations over all hours. In applying the rollback methodology in the first draft of the REA, this issue was addressed by imposing a lower bound on O₃ concentration values based on modeled O₃ levels after eliminating all U.S. anthropogenic emissions of O₃ precursors (NO_x and VOC). These estimates were developed using the GEOS-Chem global chemical transport model. Imposing a lower bound was applied so that O₃ concentrations for any particular hour would not go below the estimated U.S. background level.

For the second draft of the REA, the Agency is evaluating approaches for simulating attainment of current and alternative standards that are based on modeling the response of O₃ concentrations to reductions in anthropogenic NO_x and VOC emissions using the Higher-order Decoupled Direct Method (HDDM) capabilities in the Community Multi-scale Air Quality (CMAQ) model. This modeling incorporates estimated emissions, including emissions from non-anthropogenic sources and anthropogenic emissions from sources in and outside of the U.S. One of the expected consequences of applying the HDDM will be changes in the distribution of hourly average concentrations. The EPA notes in the REA that reducing emissions required to

meet the current standards may lead to reductions in hourly concentrations for most hours of the day, but may have little impact on concentrations for some hours, and in some cases may lead to increases in O₃ concentrations particularly during nighttime hours (US EPA, 2012a). Using actual trending data, Lefohn et al. (1998) noted that such effects occurred. They reported that as O₃ levels improved (i.e., the environment experienced lower O₃ exposures) due to reduced precursor emissions, reductions in the number of both high and low hourly average concentrations occurred. The authors also noted that as air quality improved at specific monitoring sites, the distribution of hourly average concentrations moved from both the high end as well as the low end of the distribution toward the center (i.e., 30-60 ppb) of the distribution. The reduction in the number of low hourly average O₃ concentrations was associated with lack of NO_x scavenging (US EPA, 1996).

As a result of applying the HDDM, the EPA believes that it will not be necessary to specify values for U.S. background because of the assumption that estimates of background can be obtained from the modeling directly. However, if the model does not contain modules that can adequately address natural processes that contribute to background, such as stratospheric-tropospheric exchange, estimates of U.S. background levels will likely be subject to great uncertainties, which in turn, can affect risk estimates. Therefore, the role of U.S. background deserves closer scrutiny.

On page 3-8, the current REA notes that the air quality assessment does not include estimates of background O₃, with the exception of providing a floor for O₃ concentrations when implementing the quadratic rollback method to simulate attainment of the current standards. Throughout the REA, the Agency implies that U.S. background plays an unimportant role in the risk analysis. For example, the REA notes on page 7-42 that

Given that the risk assessment focuses primarily on the reduction in risk associated with moving from recent conditions to simulated just meeting the current standard, the impact of uncertainty in U.S. background levels on the risk estimates is expected to be low, since generally, both recent conditions and current standard O₃ levels occur well above U.S. Background (for a particular day) and consequently, consideration of U.S. background does not factor into estimating the magnitude of deltas (risk reductions).

Although the Agency believes that differences between risk estimates based on recent conditions compared to just meeting the current standard may cancel out the uncertainty in the estimates of U.S. background O₃ levels, this does not appear to be correct. Table 7-10 on page 7-53 illustrates that as the rollback methodology is implemented the risk in the 25-50 ppb range increases. Similarly, the risk in the 25-50 ppb range is also high prior to the rollback implementation (Table 7-9). If U.S. background O₃ were in the 25-50 ppb range as indicated from empirical data, the *absolute* risk values in this range of concentrations would be lower and as a result, the total risk associated with (1) the recent conditions and (2) just meeting the current standard would be lower. The effect on the deltas if U.S. background O₃ were in the 25-50 ppb range would also more than likely result in considerably lower values. There is a tendency for the deltas to be higher for those cities that are predicted by the EPA to experience higher mortality under current conditions (i.e., Atlanta, Boston, Houston, Los Angeles, and New York) than the cities predicted to exhibit lower mortality (i.e., St. Louis, Baltimore, Cleveland, Denver, Detroit, Philadelphia, and Sacramento). This implies that estimates of U.S. background levels affect both the absolute as well as the delta calculations. As noted above, the expected shifts in the distribution of hourly average O₃ concentrations (i.e., changes in frequency of occurrence at both the high and low ends of the distribution) as a result of implementing the HDDM procedure, will affect the absolute values of the risk estimates.

The uncertainty associated with the estimates of background O₃ has a major role in developing confidence in the risk assessment results described in the REA in Chapter 6 (Characterization of Health Risks Based on Clinical Studies) and Chapter 7 (Characterization of Health Risk Based on Epidemiological Studies). The level of estimated U.S. background is important when assessing the reliability of the estimates of the Lowest Measured Level (LML) values described in the Health Risk and Exposure Assessment for Ozone document (US EPA, 2012a). For example, Table 7-5 on page 7-32 in the REA identifies the LML levels that are used to estimate risk using the results from the epidemiological studies. Table 7-5 reproduced below illustrates the low concentration levels used in the risk analysis. Many of these concentration levels are well below measured background O₃ concentrations.

Table 7-5 Composite Monitor O₃ LML Used in Defining Ranges of Increased Confidence in Modeling Risk

Urban Study Area	8r max (city-specific O ₃ season) ppb	8hr mean (reflects June-August levels) ppb
<i>Metrics Based on 2007 Composite Monitors</i>		
Atlanta	17	24
Baltimore	13	13
Boston	12	19
Cleveland	12	6
Denver	4	21
Detroit	13	19
Houston	6	10
Los Angeles	9	31
New York	10	10
Philadelphia	13	12
Sacramento	13	30
St. Louis	8	22
<i>Metrics Based on 2009 Composite Monitors</i>		
Atlanta	5	21
Baltimore	9	24
Boston	12	17
Cleveland	15	16
Denver	16	22
Detroit	14	11
Houston	7	15
Los Angeles	8	22
New York	8	12
Philadelphia	9	14
Sacramento	5	30
St. Louis	7	22

The "Heat Map" Tables (7-9 and 7-10) presented on page 7-53 illustrate the distribution of O₃-related all-cause mortality across distributions of daily 8-h maximum O₃ levels for each urban study area. The estimates are based on epidemiological study results. The colors in the cells reflect the size of the mortality estimates. Table 1 below summarizes O₃-related all-cause mortality across daily 8-h maximum O₃ levels between 25 to 50 ppb for each of the 12 urban study areas. Nine of the 12 cities were predicted to experience approximately 50% or more of O₃-related all-cause mortality for daily 8-h maximum O₃ levels between 25 and 50 ppb.

Table 7-9 Heat Map Table: Short-Term O₃ Exposure-Related All-Cause Mortality – Recent Conditions (2007) (Bell et al, 2004 C-R functions) (illustrates distribution of O₃-related all-cause mortality across distribution of daily 8hr max O₃ levels for each urban study area – colors in cells reflect size of mortality estimate)

Study area	Daily 8hr Max Ozone Level (ppb)																Total
	0-5	5-10	10-15	15-20	20-25	25-30	30-35	35-40	40-45	45-50	50-55	55-60	60-65	65-70	70-75	>75	
Atlanta, GA	0	0	0	0	1	3	10	13	17	22	42	51	39	40	34	52	323
Baltimore, MD	0	0	0	0	2	4	6	14	14	14	14	10	16	4	5	3	106
Boston, MA	0	0	0	0	6	20	26	32	43	39	23	33	13	26	19	26	307
Cleveland, OH	0	0	0	0	1	5	7	13	13	15	14	9	6	10	7	7	109
Denver, CO	0	0	0	0	1	1	2	3	5	6	6	6	2	1	0	0	32
Detroit, MI	0	0	0	0	1	2	6	9	13	17	5	10	6	6	5	13	94
Houston, TX	0	0	2	7	18	23	34	30	26	28	21	15	9	19	12	2	244
Los Angeles, CA	0	0	1	10	26	41	69	66	99	87	103	88	46	40	24	27	729
New York, NY	0	0	0	15	22	60	69	70	99	60	49	40	50	73	27	23	658
Philadelphia, PA	0	0	0	0	1	4	6	10	11	11	12	11	11	8	7	7	98
Sacramento, CA	0	0	0	1	3	5	9	14	14	19	17	8	8	3	4	4	110
St. Louis, MO	0	0	0	0	1	3	8	14	18	16	24	21	24	10	9	25	174

Table 7-10 Heat Map Table: Short-Term O₃ Exposure-Related All-Cause Mortality – Simulation of Meeting the Current Standard (2007) (Bell et al., 2004 C-R functions) (illustrates distribution of O₃-related all-cause mortality across distribution of daily 8hr max O₃ levels for each urban study area – colors in cells reflect size of mortality estimate)

Study area	Daily 8hr Max Ozone Level (ppb)																Total	Delta
	0-5	5-10	10-15	15-20	20-25	25-30	30-35	35-40	40-45	45-50	50-55	55-60	60-65	65-70	70-75	>75		
Atlanta, GA	0	0	0	0	2	5	12	18	21	57	43	45	29	16	6	4	260	63
Baltimore, MD	0	0	0	0	3	4	10	15	15	14	8	5	2	0	0	0	90	16
Boston, MA	0	0	0	1	8	23	24	48	26	39	32	14	26	24	7	11	282	26
Cleveland, OH	0	0	0	1	2	5	11	13	16	14	10	9	9	5	2	2	98	11
Denver, CO	0	0	0	0	1	1	3	3	7	7	5	2	0	0	0	0	30	3
Detroit, MI	0	0	0	0	1	3	7	10	17	9	9	7	7	6	6	4	86	8
Houston, TX	0	0	2	8	22	24	37	31	28	21	12	18	11	1	0	0	217	27
Los Angeles, CA	0	0	2	17	35	64	70	119	113	81	44	15	7	0	0	0	567	162
New York, NY	0	0	1	13	31	69	76	103	55	63	50	66	44	0	13	0	585	73
Philadelphia, PA	0	0	0	0	2	5	8	12	12	12	8	11	5	2	1	2	82	16
Sacramento, CA	0	0	0	1	3	7	15	14	21	13	8	5	3	0	1	0	90	20
St. Louis, MO	0	0	0	0	2	4	10	20	16	23	24	21	12	12	10	4	157	17

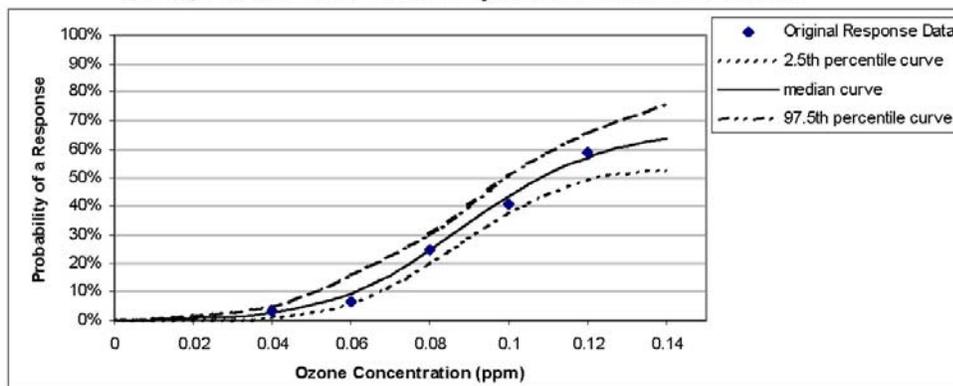
For New York City, the EPA estimates that for recent conditions that 8-h daily maximum concentrations between 25 to 50 ppb result in 358 deaths or 54% of the estimated total all-cause mortality. As will be discussed in our report, these concentration levels are in the range of background O₃ concentrations observed in the US.

Table 1. Summary O₃-related all-cause mortality across daily 8-h maximum O₃ levels of 25-50 ppb for each urban study area under recent conditions that is based on the EPA’s Heat Map analysis presented in Table 7-9.

City	O ₃ -related all-cause mortality	Percent of Total
Atlanta, GA	65	20%
Baltimore, MD	52	49%
Boston, MA	160	52%
Cleveland, OH	53	49%
Denver, CO	17	53%
Detroit, MI	47	50%
Houston, TX	141	58%
Los Angeles, CA	362	50%
New York, NY	358	54%
Philadelphia, PA	42	43%
Sacramento, CA	61	56%
St. Louis, MO	59	34%

When characterizing health risks based on clinical studies, the range of U.S. background level concentrations is also important. On page 6-12, the REA illustrates the probabilistic exposure-response relationships for FEV₁ decrement $\geq 10\%$. Figure 6-4a is reproduced below.

a) FEV₁ Decrement $\geq 10\%$
Figure 6-4. a, b, c. Probabilistic Exposure-Response Relationships for FEV₁ Decrement $\geq 10\%$, $\geq 15\%$, and $\geq 20\%$ for 8-Hour Exposures At Moderate Exertion*



Although the probability of a response below 50 ppb is low, a large fraction of the population is estimated to be exposed to these low O₃ concentrations (US EPA, 2012a). These low O₃ concentrations are used in the risk model to predict the number of persons responding.

Should the range of O₃ concentrations associated with U.S. background actually be much greater than predicted by the CMAQ model, then the risk predicted using clinical study results for the large fraction of the population may not be able to be reduced if reductions in emissions were to occur.

By limiting the discussion about the estimates of U.S. background uncertainty to the Policy Assessment (PA) document, the authors of the REA have minimized the importance of this uncertainty and the role U.S. background plays in risk estimates. By removing the focus on the uncertainty associated with estimating U.S. background hourly average O₃ concentrations, the Agency provides greater credence to the predicted clinical and epidemiological risk estimates than is deserved. It is our opinion that to develop a level of confidence in the estimated risk estimates, it is necessary to address how uncertainty in the estimate of U.S. background hourly average concentrations systematically affects the Agency's risk calculations. Without adequately discussing in the REA the uncertainties in estimating hourly average background O₃ concentrations, readers will focus on the magnitude of the absolute and difference values of the risk estimates. It is extremely important that the magnitude of the risk estimates be evaluated simultaneously with the large uncertainties associated with estimating U.S. background concentrations. The background O₃ concentrations are an important underpinning of the risk estimates.

The EPA describes some of the modeling limitations noted in McDonald-Buller et al. (2011). On page 3-63 in the ISA (US EPA, 2012b), the EPA notes that while GEOS-Chem is capable of simulating seasonal or monthly mean maximum daily average 8-h concentrations (MDA8) O₃ to within a few parts per billion on a regional basis throughout the U.S., neither the GEOS-Chem nor CAMx models are capable of simulating the 4th highest MDA8 O₃ to within

suitable bounds on a day-specific basis. The ISA (page 3-63) points out that currently all chemical transport models and not just the ones evaluated in the ISA, have difficulty in predicting day-specific quantities. Thus, based on EPA's conclusions in the ISA (US EPA, 2012b), estimates of either NA background or U.S. background O₃ concentrations over a short-time frame (e.g., hourly or 8-h average concentrations) may not be reliable on a daily basis.

In addition to the EPA ISA, McDonald-Buller et al. (2011) also point out that GEOS-Chem and other global models have difficulty representing the fine structures of O₃ events observed at relatively remote monitoring sites in the U.S., including events for which the contribution of U.S. background O₃ is likely important. McDonald-Buller et al. (2011) note that stratosphere-troposphere exchange can contribute to background O₃ at both low- and high-altitude sites. In addition, the authors note that fire plumes transported on intercontinental scales can contain very high O₃ concentrations. These plumes are generally transported in the free troposphere above the boundary layer, and have a strongly layered structure that is difficult to capture with Eulerian models. McDonald-Buller et al. (2011) note that observational data are essential to (a) validate models and improve the confidence in their performance, (b) better understand the causes of enhanced hourly averaged O₃ concentrations ≥ 50 ppb), (c) indicate geographic areas of strength and weaknesses, and (d) guide model improvements where needed. The purpose of this report is to place into perspective the amount of uncertainty in the Agency's estimate of hourly average background O₃ concentrations.

2. The Importance of the Stratosphere Contributing to Surface Ozone Concentrations

The estimate of natural background O₃ concentrations plays an important role in estimating NA and U.S. background O₃ concentrations. As noted in the ISA (US EPA, 2012b),

natural background O₃ concentrations are affected by emissions from sources, such as stratospheric intrusion, wildfires, and lightning. In this section we discuss the importance of the stratosphere contribution to the enhancement of hourly average O₃ concentrations (i.e., hourly average concentrations ≥ 50 ppb). The REA notes on page 2-3 that contribution to O₃ concentrations in an area from the stratosphere are defined as being part of background O₃.

Evidence exists that frequently occurring stratosphere-to-troposphere transport to the surface (STT-S) processes are affecting surface hourly average background O₃ concentrations at both high- and low-elevation monitoring sites during the springtime, as well as other times during the year (Ludwig et al., 1977; Haagenson et al., 1981; Davies and Schuepbach, 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; Lefohn et al., 2011; Ambrose et al., 2011; Cooper et al., 2011; Langford et al., 2011; Lefohn et al., 2012-submitted; Lin et al., 2012-submitted).

Findings published in the literature suggest that background O₃ makes a substantial contribution to O₃ levels under a variety of meteorological conditions – both relatively clean conditions and those where local photochemical production contributes to the exceedance of air quality standards. Lefohn et al. (2001) attributed stratospheric-tropospheric exchange (STE) processes to the observation that hourly average O₃ concentrations ≥ 50 and 60 ppb occurred frequently during the photochemically quiescent months in the winter and spring at several rural high- and low-elevation monitoring sites across southern Canada and the northern United States. The authors noted that high-elevation (e.g., Yellowstone National Park-2468 m in Wyoming) as well as lower elevation sites in the US and Canada (e.g., Voyageurs National Park in Minnesota) frequently experienced enhanced (i.e., ≥ 50 ppb) hourly average O₃ concentrations during the

springtime across multiple years. Confirming the suggestion by Lefohn et al. (2001) that STE processes can contribute to violations of the US 8-h O₃ standard, Langford et al. (2009) documented STE episodes that resulted in exceedances of the US O₃ standard during the springtime.

Using the Lagrangian Analysis Tool (LAGRANTO) trajectory model (Wernli and Davies, 1997), Lefohn et al. (2011) investigated the frequency of STE events and their associated enhancements at 12 surface O₃ monitoring sites in the western and northern tier of the US, including the site at Yellowstone National Park. For most of the sites analyzed, Lefohn et al. (2011) indicated that the STT-S contributions were frequent during specific months and appeared to enhance the surface O₃ concentrations at both high- and low-elevation monitoring sites. For the sites characterized, the authors noted that Yellowstone exhibited the greatest coincidences during the spring and summer for daily maximum hourly average O₃ concentration ≥ 50 ppb with stratosphere-to-troposphere transport to the surface. During the months of March, April, and May of 2006, Yellowstone NP experienced 1,240 hourly average O₃ concentrations ≥ 50 ppb. Using data from this site, Lefohn et al. (2011) reported that for those months in which statistically significant coincidences occurred, the daily maximum hourly springtime average O₃ concentrations were usually in the 60 to 70 ppb range; the maximum daily 8-h average concentrations mostly ranged from 50 to 65 ppb. At many of the lower elevation sites, there was a preference for O₃ enhancements to be coincident with STT-S during the springtime, although summertime occurrences were sometimes observed. For those months in which statistically significant coincidences occurred, the daily maximum hourly average concentrations were mostly in the 50 to 65 ppb range; the daily maximum 8-h average concentrations were usually in the 50 to 62 ppb range.

Lefohn et al. (2012-submitted) quantified the frequency of STE events that resulted in O₃ concentration enhancements (i.e., hourly average concentrations \geq 50 ppb) observed at 39 high- and low-elevation monitoring sites in the US for the years 2007-2009. They employed a refined forward trajectory-based approach to address the relationship between stratospheric intrusions and enhancements in hourly average O₃ concentrations. The results indicated that STT down to the surface (STT-S) frequently contributed to enhanced surface O₃ hourly averaged concentrations at sites across the US, with substantial year-to-year variability. Months with a statistically significant coincidence between enhanced O₃ 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. However, besides the high-elevation sites, low-elevation monitoring sites across the entire US experienced enhanced O₃ concentrations that were coincident with STT-S events. Examples of sites with a statistically significant coincidence between STT-S and enhanced O₃ concentrations were Voyageurs NP (MN), Chittenden County (VT), Yellowstone NP (WY), Lassen Volcanic NP (CA), Crestline (CA), El Dorado County (CA), Canyonlands NP (UT), Jefferson County (CO), Mesa Verde NP (CO), Harris County (TX), Shenandoah NP (VA), Cuyahoga (OH), and Georgia Station (GA).

The Agency in its ISA (US EPA, 2012b) notes on page 3-34 that Lefohn et al. (2011) identified likely stratospheric influence at the surface sites on a number of days during the spring of 2006 to 2008. The EPA noted that while the Lefohn et al. (2011) analysis captured the frequency and vertical penetration of stratospheric intrusions, the authors did not provide information about the contribution of the intrusions to the measured O₃ concentrations. To assist the Agency in its assessment of the science, we point out that Lefohn et al. (2012-submitted)

provide information that quantifies the concentration enhancements associated with STE. Of specific relevance to the EPA and ISA (US EPA, 2012b), Lefohn et al. (2012-submitted) provided as part of their analyses an evaluation of data for those sites that were least affected by days in which zero STT-S counts coincided with enhanced O₃ concentrations (i.e., ≥ 50 ppb). In other words, these O₃ monitoring sites did not appear to be influenced during specific months by lightning, long-range transport from Asia, wildfires, or anthropogenic perturbations during the months that showed statistically significant coincidences between STT-S and enhanced O₃ concentrations. Numerous days were experienced when enhanced O₃ concentrations occurred. The high-elevation sites experienced a greater percentage of days with concentrations ≥ 50 ppb and ≥ 60 ppb on days when STT-S >0 during the springtime. For example, over the spring period for those days in which the STT-S >0 , the high-elevation Yellowstone NP (WY) site experienced approximately 86% and 72% of the total days with 1-h and 8-h daily maximum concentrations ≥ 50 ppb, respectively. For Lassen Volcanic NP (CA), approximately 82% and 62% of the days exhibited 1-h and 8-h daily maximum concentrations ≥ 50 ppb, respectively. In comparison, the low-elevation sites (e.g., Voyageurs NP, Cuyahoga County, and Chittenden County) that were least affected by days in which zero STT-S counts coincided with enhanced O₃ concentrations (i.e., ≥ 50 ppb), experienced lower percentages in the spring than the higher-elevation locations when STT-S >0 . Voyageurs NP experienced approximately 66% and 44% of the days with 1-h and 8-h daily maximum concentrations ≥ 50 ppb, respectively, while Cuyahoga County experienced approximately 64% and 45% of the days with 1-h and 8-h daily maximum concentrations ≥ 50 ppb, respectively. Chittenden County had approximately 66% and 51% of the days with 1-h and 8-h daily maximum concentrations ≥ 50 ppb, respectively.

Lefohn et al. (2012-submitted) reported that there was no distinction between the high- and low-elevation sites for the 1-h and 8-h daily maximum concentrations ≥ 70 ppb when $STT-S > 0$ during the springtime. Maximum concentrations showed little if any dependence on altitude. For Yellowstone NP and Lassen Volcanic NP, the highest 8-h daily maximum concentration during the springtime was 68 ppb and 73 ppb, respectively. The highest 8-h daily maximum concentrations experienced during the springtime at Voyageurs NP, Cuyahoga County, and Chittenden County were 63, 77, and 83 ppb, respectively.

Papers by Cooper et al. (2011), Langford et al. (2011), McDonald-Buller et al. (2011), and Lin et al. (2012-submitted) provide evidence for the role of elevated background O_3 concentrations in O_3 events exceeding or approaching air quality standards. Langford et al. (2011) found that surface O_3 measurements from 41 sampling stations indicate that $\sim 13\%$ of the variance in the maximum daily 8-h average (MDA8) O_3 between May 10 and June 19, 2010 was associated with changes of 2-3 day duration linked to the passage of upper-level troughs. Ozonesondes launched from Joshua Tree National Park and airborne lidar measurements show that these changes coincided with the appearance of stratospheric intrusions in the lower troposphere above southern California. In one event, the intrusion led to a peak 1-h O_3 concentration of 88 ppb at Joshua Tree National Monument near the ozonesonde launch site and widespread entrainment of stratospheric air into the boundary layer that increased the local background O_3 over the entire area to ~ 55 ppb. This background was 10-15 ppb higher than the baseline O_3 in air transported ashore from the Pacific Ocean, and when combined with locally produced O_3 , led to several exceedances of the current National Ambient Air Quality Standard (NAAQS) on the following day.

Cooper et al. (2011) investigated the contribution to baseline O₃ concentrations in California over a 6-week period (May 10 – June 19, 2010). The authors found that on average approximately 8-10% of the baseline O₃ that enters California in the 0-6 km range impacts the surface of the USA. Within California, the major impact of baseline O₃ above 2 km is on the high-elevation terrain of eastern California. Baseline O₃ below 2 km has its strongest impact on the low-elevation sites throughout the state. Cooper et al. (2011) indicated that descending stratospheric intrusions and Asian pollution plumes played an important influence on the O₃ concentration distributions along the California coast. The authors indicated that the influences associated with descending stratospheric intrusions and Asian pollution plumes were not mutually exclusive as most of the pollution plumes identified were combined with air masses with varying degrees of stratospheric origin. Over the 6-week investigation, the greatest enhancements to surface O₃ from baseline sources occurred over the Los Angeles Basin (32-63%).

Ambrose et al. (2011) investigated the importance of STE events versus long-range transport affecting lower tropospheric O₃ concentrations at the Mt. Bachelor Observatory (2763 m) in central Oregon. A total of 25 high-O₃ events, defined as time periods when the 8-h averaged O₃ was >70 ppb, were recorded. The events occurred between early March and late September 2004-2009. For the total number of classifiable high O₃ event hours, the authors reported that the enhanced O₃ levels in the free troposphere were transported to the high-elevation site and were driven mostly by subsidence of O₃-rich air masses from the upper troposphere/lower stratosphere (~52%), Asian long-range transport (~13%), and a combination of Asian long-range transport and upper troposphere/lower stratosphere influences (~36%). Ambrose et al. (2011) concluded that downward mixing of O₃-rich air masses from the upper

troposphere/lower stratosphere together with trans-Pacific transport of urban/industrial and biomass burning emissions from the Asian continent were the most important mechanisms for delivering enhanced O₃ levels to the lower free troposphere in the Pacific Northwest.

Based on high resolution runs of the NOAA Geophysical Fluid Dynamics Laboratory AM3 model during the 2010 CalNex campaign, Lin et al. (2012-submitted) have reported that the stratospheric contribution to the North American background (NAB) in the spring is larger than that from Asian transport (Fig. 1). 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. This feature enables a process-oriented analysis for the intermingling of Asian pollution with stratospheric air over the west coast of the US (Lin et al., 2012). Air with stratospheric enhancements is regularly found to be interleaved with layers of Asian pollution with elevated O₃ (Cooper et al., 2004; Ambrose et al., 2011; Lin et al., 2012). Lin et al. (2012-submitted) reported that the background contribution is largest in the 50-80 ppb range of daily 8-h maximum averages (MDA8), which has implications for meeting air quality standards, as well as affecting human health risk estimates associated with establishing O₃ standards.

If the results of the Lin et al. (2012-submitted) study for the late spring/early summer period in a single year (2010) are representative of this time of year, the background O₃ contribution from this study appears to be somewhat larger than the results in Zhang et al. (2011) cited in the latest version of the ISA (US EPA, 2012b), especially for the stratospheric contribution. This may partially account for the bias between the measured and modeled seasonal tropospheric O₃ at Trinidad Head reported by McDonald-Buller et al. (2011).

Recent studies reported by investigators indicate that modeling estimates of NA background may be underestimated. Emery et al. (2012) used both a low-resolution GEOS-Chem global model and a high-resolution regional (12 km) chemical transport model in estimating NA background. The authors compared differences in model predictive performance against O₃ observations and differences in temporal and spatial background variability over the US. Regional modeling over the North American continent was conducted using the Comprehensive Air quality Model with extensions (CAMx). Emery et al. (2012) reported that in general the CAMx performed better in replicating observations at remote monitoring sites, and performance remained better at higher concentrations. While spring and summer mean NA background predicted by GEOS-Chem ranged 20-45 ppb, CAMx predicted NA background ranged 25-50 ppb and reached well over 60 ppb in the West and Intermountain West due to event-oriented phenomena, such as stratospheric intrusion and wildfires. CAMx showed a higher correlation between modeled NA background and total observed O₃. A case study during April 2006 suggested that stratospheric exchange of O₃ was underestimated in both models on an event basis. The authors concluded that wildfires, lightning NO_x, and stratospheric intrusions contribute a significant level of uncertainty in estimating NA background.

Using a slightly modified model from Emery et al. (2012), Lefohn, Emery, and co-investigators further explored the performance of the GEOS-Chem/CAMx for estimating NA background O₃ concentrations, as well as total O₃. The GEOS-Chem/CAMx runs used the same 2006 CAMx datasets as described in Emery et al. (2012). The model run invoked the CAMx Ozone Source Apportionment Technology (OSAT) to track contributions of boundary conditions and natural sources to total hourly O₃ throughout the entire modeling domain. The authors combined the data analysis techniques described in Lefohn et al. (2011; 2012 submitted) with the

2006 annual GEOS-Chem/CAMx modeling approach described by Emery et al. (2012) to investigate the relationships between diagnosed stratosphere-troposphere transport to the surface (STT-S) events and uncertainties associated with model estimates of NA background O₃ on a daily basis. Source apportionment modeling specifically tracking STE, regional transport from Asia, and natural source contributions allowed the investigators to break out the contributions to simulated NA background.

Modeling results were evaluated against hourly observation data at 8 sites and analyzed with respect to STT-S events as diagnosed by Lagrangian trajectory modeling (Lefohn et al., 2011; 2012-submitted). Lefohn, Emery, and co-investigators concluded that the modeling system was highly dependent on the proper specification of boundary conditions derived from GEOS-Chem, particularly in the western US, where CAMx under predicted total O₃ at rural sites in the spring through early summer seasons when little anthropogenic contribution was evident. Fig. 2 illustrates the monthly median bias and STT-S counts at Yellowstone NP for 2006. Months with statistically significant STT-S coincidences with enhanced O₃ are indicated with an asterisk (*). The figure shows that hourly O₃ during the late winter, spring, and early summer months tended to be underestimated, and this also was the period when STT-S counts were the highest. The monthly coincidences between enhanced (i.e., ≥ 50 ppb) O₃ concentrations and STT-S events were also statistically significant during this period of time. Model median bias during the summer months was fairly small with November and December experiencing similar bias values as those experienced during the late spring/early summer months. Fig. 2 suggests that months with the largest statistically significant STT-S coincidences were also the months with the largest negative median bias at Yellowstone NP. This indicates that the model may not have predicted the full impact of these STT-S events on surface O₃ during the spring, although other sources of

error, including underestimates of long-range tropospheric transport, need to be considered. Fig. 3 shows the GEOS-Chem time series for Yellowstone NP that was presented in the ISA. GEOS-Chem is under predicting the observed values.

Lefohn, Emery, and co-investigators reported that at sites influenced by anthropogenic sources, where chemistry involving local emissions decays and obscured the background O₃ signal in the model, STT-S analyses provided supplemental information on explaining enhancements of the O₃ concentrations (Fig. 4). The results associated with the investigation by Lefohn, Emery, and co-workers appear to agree with the conclusions reached by Emery et al. (2012) and the EPA (2012b) that annual coarse-resolution modeling applications have difficulty in characterizing observed O₃ concentrations on a short-time frame, such as for the daily time series of hourly or daily 8-h average O₃ concentrations.

3. Estimating North American Background Ozone Concentrations Using Empirical Data

3.1 The Influence of Background O₃ Concentrations at Trinidad Head (CA)

McDonald-Buller et al. (2011) note that PRB O₃ (now referred to as NA background by the EPA) is a model construct that must be informed by and evaluated based on observational data. Similarly, the model-constructed U.S. background O₃ that the Agency uses for its risk assessments must also be informed by and evaluated based on observational data.

The definitions of background discussed in the Introduction have been formulated in the context of model calculations and it is assumed and not directly measurable quantities. Recent work discussed in McDonald-Buller et al. (2011) concludes that conditions representative of background O₃ are routinely encountered at selected monitoring sites. Two sites noted in this publication that are particularly likely to observe O₃ under conditions representative of

background are Trinidad Head, California and Mt. Bachelor, Oregon. While a contribution from North American emissions to the hemispheric background is also included in the measured O₃ concentrations at these representative monitoring locations, this contribution is likely to be small (Zhang et al., 2011). One of these sites, Trinidad Head, California, is situated on a large domed prominence to the west of the town of Trinidad, which is a small town of about 400 people on California's north coast. The site is located at 124.1° W and 41.1° N at an elevation of 107m (Fig. 5). The site is connected to the mainland only on its northern end. Surface O₃ measurements began in mid-April 2002 and continue to the present at Trinidad Head. In addition O₃ vertical profiles using ozonesondes have been measured from a nearby site (the town of Trinidad) since 1997. Ozone soundings are performed routinely on a weekly basis, but have been done in campaign mode where daily soundings have been conducted for periods of 4-6 weeks during the spring and summer (Oltmans et al., 2008; Cooper et al., 2006, 2007, 2011). There is a prominent seasonal cycle in O₃ throughout the troposphere at Trinidad with a maximum in April and May (Figs. 6 and 7). Long-range transport outside of North America and natural processes such as stratospheric enhancement contribute to O₃ concentrations measured at this site (Cooper et al., 2011). Trinidad Head, CA, experiences its airflow pattern overwhelmingly from the North Pacific Ocean during all seasons with stronger flow during the winter and spring months that regularly meet background conditions. The frequency of hourly average concentrations ≥ 50 ppb in the springtime (e.g., month of April), when almost all of the high concentrations occur, is large (Table 2) and varies from year to year (i.e., 30 to 187 hours).

Table 2. Summary of the number of hourly average concentrations ≥ 0.05 ppm and maximum hourly average value (in parentheses) measured at Trinidad Head, California for the 10-year period, 2002 through 2011. UTC time period.

Year	March	April	May	June	July
2002	No Data	36 (54 ppb)	18 (53 ppb)	0 (41 ppb)	0 (45 ppb)
2003	23 (52 ppb)	187 (64 ppb)	96 (57 ppb)	13 (54 ppb)	0 (41 ppb)
2004	38 (62 ppb)	30 (54 ppb)	3 (50 ppb)	2 (50 ppb)	0 (38 ppb)
2005	58 (55 ppb)	72 (56 ppb)	6 (52 ppb)	0 (46 ppb)	0 (39 ppb)
2006	37 (55 ppb)	56 (60 ppb)	43 (64 ppb)	0 (47 ppb)	0 (35 ppb)
2007	21 (52 ppb)	46 (57 ppb)	30 (55 ppb)	0 (40 ppb)	0 (33 ppb)
2008	26 (53 ppb)	120 (61 ppb)	34 (56 ppb)	3 (52 ppb)	0 (42 ppb)
2009	18 (54 ppb)	73 (65 ppb)	11 (54 ppb)	0 (43 ppb)	0 (35 ppb)
2010	38 (58 ppb)	51 (58 ppb)	6 (52 ppb)	0 (42 ppb)	0 (32 ppb)
2011	18 (54 ppb)	33 (57 ppb)	5 (51 ppb)	0 (48 ppb)	0 (30 ppb)

The range of maximum hourly average concentrations for the month of April over the period 2002-2011 is 54 – 65 ppb. For the period 2003-2011, daily maximum 8-h average O₃ concentrations exceeded 60 ppb during the springtime. The 3-year average of the 4th highest maximum 8-h average concentrations fell within the range of 50 to 52 ppb. These are relatively high values at a marine boundary layer location under conditions representative of background O₃ occur during the springtime. That Trinidad Head regularly makes measurements under background O₃ conditions for daytime observations (mid morning to late afternoon) has now been demonstrated by a number of studies (Oltmans et al., 2008; Goldstein et al., 2004; Parrish et al., 2010; Cooper et al., 2011; Huang et al., 2010). Oltmans et al., (2008) used both a trajectory climatology (cluster analysis) (Fig. 8), as well as individual trajectories for O₃ events (Fig. 9), where O₃ hourly averages were ≥ 50 ppb to show that the broad scale flow influencing Trinidad Head was overwhelming from off the Pacific Ocean and thus represented background conditions. In addition they analyzed the local wind conditions for each O₃ hourly average (Fig. 10) and

showed that high O₃ amounts are associated with local winds off the ocean and occur primarily during the daytime. At night on the other hand the Trinidad Head site experiences flow from off the land that shows the influence of O₃ deposition and possible chemical loss. The study of Goldstein et al. (2004) that made detailed chemical measurements during a campaign in April and May 2002 used several tracers to separate local influence from recently polluted air and thus filter out significant influences from North American continental emissions. Continental influence was most often associated with nighttime observations coming with the offshore (from the land) winds. Under these circumstances the O₃ concentrations would not be representative of background O₃ conditions and have lower O₃ amounts (see Fig. 10) as noted by Goldstein et al. (2004), Parrish et al. (2009), and Cooper et al. (2011). The observations at Trinidad Head can be screened for flow off the ocean versus from over land to determine the diurnal cycle (Oltmans et al., 2008) under background O₃ conditions (Fig. 11). The diurnal pattern at Trinidad Head deduced from these observations suggest that even at night average background O₃ values in the spring are over 35 ppb. During the summer when Trinidad Head is primarily influenced by marine boundary layer air, the nighttime average for background O₃ conditions is nearly 25 ppb.

Using the FLEXPART Lagrangian particle model, Cooper et al. (2011) screened the portion of the ozonesonde profiles below 3 km during IONS 2010/CalNex for North American influence and found slightly lower O₃ in profiles at the three coastal site in northern California, when the profiles were influenced by North American emissions, although these differences were not significant (Fig 12). Cooper et al. (2011) concluded that the baseline sites of Trinidad Head (TH), Point Reyes (RY), Point Sur (PS), and San Nichols Island (SN), all situated on the coast, have very low exposure to local emissions and mid-afternoon exposure to marine air masses. By calculating retroplumes using the FLEXPART model for each ozonesonde profile it was determined that the air masses travel across the North Pacific Ocean, with the more northern sites having

a greater influence from high latitude regions, and the more southern sites having more influence from lower latitude regions (Cooper et al., 2011). Based on this, they concluded that the median O₃ profiles above these sites are representative of baseline O₃ along the U.S. west coast (Fig. 13). In the lower troposphere, the median O₃ values at these sites are very similar indicating that the O₃ distribution is dominated by the broad scale flow off the Pacific Ocean. This strong similarity in lower troposphere O₃ among all of the sites suggests that the O₃ values at Trinidad Head are indicative of O₃ amounts reaching the California coast from the Pacific.

Parrish et al. (2010) showed that CO measured in aircraft flask samples off the coast at Trinidad Head agrees well with the global background CO concentration determined from the NOAA ESRL Global Monitoring Division flask network for the latitude, season and years of the aircraft CO measurements (Fig. 14). This indicates that the air sampled in the troposphere just upwind of Trinidad Head does represent background northern mid-latitude air, without discernable direct CO influence from North America. In photochemically well-processed air masses in summertime, any O₃ enhancements from relatively local continental sources are expected to be only a fraction of the CO enhancements. Thus, the air sampled represents background northern mid-latitude air with respect to O₃ as well as CO.

All drafts of the ISA, including the current one (June 2012), have misrepresented the frequency with which observations of background O₃ are measured at Trinidad Head. The term “at times” is used to characterize the frequency that the site at Trinidad Head intercepts air from off the Pacific Ocean. The ISA suggests that the value of 30% used by Parrish et al. (2009) for their analysis is representative of conditions representative of background O₃. The work of Goldstein et al. (2004) and Oltmans et al. (2008) conclude that background O₃ conditions are observed much more frequently. Goldstein et al. (2004) state

A regular diurnal meteorological pattern occurred ... at Trinidad Head with strong daytime winds out of the north-west (off the ocean), and weaker and more variable winds at night. As a result, air sampled during the day was typically of marine origin with little recent continental influence, whereas at night the effects of recent continental influence were commonly observed. The distribution of observations for wind speed and direction... emphasizes that winds out of the north-west were the dominant meteorological feature. Ozone concentrations had an inverse pattern of concentration variations, suggesting that when local pollution sources were observed, ozone was depleted by surface deposition or chemical reactions. Under the dominant daytime meteorological pattern, strong winds out of the north-west, the concentrations of CO, CO₂, and MTBE were not enhanced, nor was the concentration of O₃ depleted. Filtering out local influences removed 20 to 40% of the observations, depending on the constraints applied, decreased the mean CO mixing ratio by 5%, and increased the mean O₃ mixing ratio by 8%.

Detailed analysis of surface and profile O₃ measurements along the California coast demonstrate that these sites are dominated by air from off the Pacific Ocean that is representative of conditions defined for background. In the spring at a marine boundary layer site, such as Trinidad Head, hourly average O₃ concentrations are often ≥ 50 ppb and in April the monthly average daytime maximum is ~ 45 ppb. Although marine boundary layer O₃ decreases through the summer, air flowing into California above the boundary layer remains on average above 50 ppb through the summer. *We conclude that there is little evidence that recent North American emissions contribute significantly to O₃ measured at coastal sites, such as Trinidad Head. In fact, such emissions usually depress O₃ levels. Between 60-80% of the time observations in the spring at Trinidad Head are free of local influences. Daytime O₃ values that overwhelmingly determine the MDA8 are almost exclusively of marine origin with little North American continental influence.*

At Mt. Bachelor, Oregon, the local wind regime is subject to orographic upslope/downslope flow with downslope flow dominating at night. Under nighttime conditions, air representative of the free troposphere is consistently sampled (Ambrose et al., 2011; Weiss-

Penzias et al., 2006). In addition, a number of tracers of both local and remote sources have been used to identify data representative of background conditions. As with O₃ of marine origin at Trinidad Head, air representative of free troposphere conditions has higher average O₃ mixing ratios than air with recent North American continental influence (Ambrose et al., 2011).

Measurements of background O₃ from sites such as Trinidad Head and Mt. Bachelor provide an important tool for assessing model performance, particularly when the models are used to address parameters such as NA background or U.S. background (McDonald-Buller, 2011). *The observed O₃ values provide a picture of the full range of O₃ concentrations that includes episodic events, especially at the higher end of the distribution that are associated with stratospheric intrusions and biomass burning. These events may not be consistently captured in models* (McDonald-Buller, 2011).

In summary, Trinidad Head overwhelmingly makes measurements under background O₃ conditions for daytime observations (mid morning to late afternoon). As noted above, the frequency of hourly average concentrations ≥ 50 ppb in the springtime, when almost all of the high concentrations occur, is large (Table 2) and varies from year to year during the month of April (e.g., 30 to 187 hours). The range of maximum hourly average concentrations for the month of April over the period 2002-2011 is 54 – 65 ppb. In the period 2003-2011, daily maximum 8-h average O₃ concentrations exceeded 60 ppb during the springtime. A 3-year average of the 4th highest maximum 8-h average concentrations is in the range from 50 to 52 ppb. When compared to the observational data at Trinidad Head (Figs. 15a and 15b), GEOS-Chem model predictions for the spring (US EPA, 2012b) indicate that daily maximum 8-h average O₃ concentrations are consistently underestimated, which suggests that model background levels are estimated to be too low for this season.

These relatively high values during the spring are observed under background O₃ conditions and should be used to correct the underestimates that are associated with the model results in establishing the range of U.S. background O₃ levels. Given the publication of several recent studies, the recommendation contained in McDonald-Buller et al. (2011) emphasizing the application of the full diversity of models for understanding the role of background O₃ for the standard-setting process appears feasible. Continued extensive evaluation of chemical transport models' ability to provide background O₃ estimates by using surface, sonde, aircraft, and satellite O₃ observations to assess the strengths and weaknesses of key processes is also a necessary requirement (McDonald-Buller et al., 2011).

3.2 Long-Range Transport Effects on NA and U.S. Background in the Western US from Eurasian Biomass Burning

Eurasian emissions associated with biomass burning and their easterly transport contribute to O₃ concentrations at west coast O₃ monitoring sites under NA and U.S. background conditions, as well as inland O₃ monitoring sites in Montana, Wyoming, and North Dakota (Oltmans et al., 2010). Published information indicates that biomass burning events in Eurasia are not infrequent. The effects of Eurasian biomass burning in producing O₃ enhancements in surface O₃ in the western US has been reported in the literature (Jaffe et al., 2004; Bertschi et al., 2004; Bertschi and Jaffe, 2005; Pfister et al., 2010; Oltmans et al., 2010). The year 2008 was one in which large spring Eurasian biomass burning occurred. Unusually high O₃ readings were recorded in April 2008 at surface O₃ monitoring sites from northern Alaska to northern California as well as inland monitoring sites in Montana, Wyoming, and North Dakota. 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. At Trinidad Head, hourly O₃ readings were

>50 ppb almost continuously for a 35-h period. At several sites in northern California, located to the east of Trinidad Head, numerous occurrences of O₃ readings exceeding 50 ppb were experienced during this period. As the biomass burning enhanced O₃ plume moved further into the interior of the US between 18-20 April through a northern tier of states (Montana, Wyoming, North Dakota), surface O₃ measurements at several monitoring sites appeared to have intercepted the plume (Oltmans et al., 2010). Trajectories from each site suggest that the enhancements observed during this period could have come from the burning region. The 8-h average O₃ enhancements were above the normal background concentrations observed at these monitoring sites (i.e., 45-55 ppb for Montana and North Dakota and 50-60 ppb for Wyoming). The 8-h daily maximum at Yellowstone on 19 April (69 ppb) suggests an enhancement during the period of suspected plume influence of 5-10 ppb above the other relatively high values observed at this site. This is also about the amount of the perturbation seen at the other interior monitoring sites (Oltmans et al., 2010). At Trinidad Head in April 2008, the occurrences of hourly averaged O₃ concentrations \geq 50 ppb were similar in magnitude to the number of events in April 2003, which over a 9-yr period experienced the highest occurrences of hourly average concentrations \geq 50 ppb. Although a thorough study of 2003 was not undertaken, modeling of 2003 data found that biomass burning impacted the west coast of North America (Pfister et al., 2010) and may have been the cause of the elevated surface O₃ amounts at Trinidad Head in April 2003 as well (Oltmans et al., 2010).

3.3 The Frequency of STE in Influencing Surface Ozone Concentrations

A key aspect that affects the importance of STE processes in replenishing NA background is the frequency of STE events. While it is recognized that STE events contribute to

the variability of natural background, it is important to quantify the frequency of those STE processes that perturb natural background O₃ concentrations that result in enhancements to observed surface O₃ levels. Some researchers report from modeling runs that the stratospheric contribution to surface O₃ levels is of minor importance (Fiore et al., 2003). On the other hand, other modelers report that the stratospheric exchange of O₃ appears to be underestimated in both regional and global modeling estimates on an event basis (Emery et al., 2012).

Indications of the importance of the frequency of STE events affecting lower tropospheric O₃ concentrations have been reported (Ordóñez et al., 2007). Reed (1955) and Danielsen (1968) used instrumented high-altitude aircraft measurements of radioactivity and O₃ to document the exchange of air and trace constituents from the stratosphere into the troposphere. Building upon this research methodology, Ludwig et al. (1977) also examined the behavior of a surrogate for stratospheric O₃ by characterizing the behavior of radioactive debris injected into the stratosphere during nuclear weapons testing in the 1960s. Using ⁹⁰Sr, Ludwig et al. (1977) concluded that a significant stratospheric contribution to ground level O₃ concentrations in the middle latitudes of the Northern Hemisphere, with a maximum occurring in the springtime, was evident at both high- and low-elevation surface-monitoring sites.

Lefohn et al. (2011) explored the frequency of STT events that were associated with O₃ concentration enhancements at 12 surface O₃ monitoring sites in the western and northern tier of the US. For most of the sites analyzed, Lefohn et al. (2011) indicated that the STT-S contributions were frequent during specific months and appeared to enhance the surface O₃ concentrations at both high- and low-elevation monitoring sites. The methodology described in Lefohn et al. (2011) has been applied by Lefohn et al. (2012-submitted) to 39 O₃ monitoring sites. Fig. 16 identifies the location of the 39 monitoring sites. The seasonal and spatial patterns

observed by Lefohn et al. (2012-submitted) for the occurrences of days and months in which O₃ concentration enhancements were statistically associated with STT-S events were similar to those reported by other investigators. Fig. 17 illustrates the average number of days per month when STT-S was coincident with O₃ enhancements by geographic region for spring (March, April, and May), summer (June, July, and August), fall (September, October, and November), and winter (December, January, and February). The numbers within each bar are the number of site-months contributing to the average. Ambrose et al. (2011) noted that for the high-elevation O₃ monitoring site at Mt. Bachelor Observatory in Oregon that enhanced 8-h average O₃ concentrations >70.0 ppb occurred during the spring, summer, and fall. The authors attributed the enhanced levels to the transport of O₃ from the upper troposphere/lower stratosphere as well as from Asia. In the West, during the spring, summer and fall, Lefohn et al. (2012-submitted) found for the high-elevation sites at Lassen Volcanic NP and Yosemite NP (Turtleback Dome) numerous days when statistically significant relationships occurred between STT-S and hourly averaged enhanced O₃ concentrations (i.e., ≥ 50 ppb).

Lefohn et al. (2011) noted the importance of STT events enhancing O₃ concentrations in the Intermountain West at Yellowstone NP, as well as other locations across the northern tier of the US. Emery et al. (2012) noted the importance of STT processes in influencing enhanced O₃ concentrations at monitoring sites in the West and Intermountain West in the US. Lefohn et al. (2012-submitted) indicate that the high-elevation sites in the West, Intermountain West, and East experience O₃ concentration enhancements frequently occurring during the spring, summer, and fall that are statistically associated with STT-S events. For the low-elevation sites in the Upper Midwest (Theodore Roosevelt NP, Voyageurs, NP, and Ann Arbor), frequent occurrences in which O₃ concentration enhancements were statistically associated with STT-S events were

observed. This pattern corresponds with the pattern described by Ludwig et al. (1977). In the Midwest and East, Lefohn et al. (2012-submitted) observed for the low-elevation sites that the occurrences of O₃ concentration enhancements statistically associated with STT-S events were less frequent than exhibited in the Intermountain West. In their analysis, Lefohn et al. (2012-submitted) observed frequent O₃ concentration enhancements related with STT-S events at the Harris County, Shenandoah NP, Rockdale, and Georgia Station sites. At the Shenandoah NP site, the frequent occurrences of enhancements to STT-S were similar in number to those observed at the higher-elevation monitoring sites at Whiteface Mountain (NY) and Mount Washington (NH). Although the authors observed less frequent O₃ enhancements associated with STT-S in the East compared to the Intermountain West and the West, low-elevation sites, such as Chittenden County (VT), exhibited O₃ concentration enhancements statistically associated with STT-S events frequently during the spring.

The Crestline site, located in southern California, is heavily influenced by anthropogenic sources. The site experiences some of the highest O₃ exposures in the US. For example in 2008-2010, the site experienced the highest 3-year average of the 4th highest daily maximum 8-h average O₃ concentration (i.e., 112 ppb) in the US (US EPA, 2009). Lefohn et al. (2012-submitted) found that the Crestline site frequently experiences enhanced O₃ concentrations statistically associated with STT-S events during the spring and fall months. Langford et al. (2011) investigated the stratospheric influence on surface O₃ in the Los Angeles area during late spring and early summer of 2010. The authors presented evidence that at times during the 43-day (May 9 to June 20, 2010) period, STT processes appeared to enhance surface O₃ concentrations at the Crestline site. Ozone concentrations associated with STT-S events appear to supplement the enhanced O₃ concentrations that are associated with locally generated anthropogenic sources.

4. Comparisons of Background Ozone Concentrations with Predicted U.S. Background Concentrations

As indicated in the previous sections, we conclude that between 60-80% of the time observations in the spring at Trinidad Head are free of local influences and that daytime O₃ concentrations that overwhelmingly determine the MDA8 are almost exclusively of marine origin with little North American continental influence. The range of maximum hourly average O₃ concentrations for April over the period 2002-2011 is 54 – 65 ppb. For the period 2003-2011, daily maximum 8-h average O₃ concentrations at times exceed 60 ppb during the springtime. The range of 3-year average of the 4th highest maximum 8-h average concentrations is from 50 to 52 ppb. The O₃ values at Trinidad Head we believe are indicative of O₃ amounts reaching the California coast from the Pacific. Fig. 18 illustrates the diurnal profiles of seasonally averaged hourly U.S. background floor values estimated by the EPA for Los Angeles and Sacramento. Profiles for spring, summer, autumn, and winter are colored in green, red, gray, and blue, respectively. The REA notes on page 4-8 that the distribution of the U.S. background floor values varied across the 12 areas studied in the risk analysis, but the values generally ranged from near 0 to between 30 and 40 ppb, with median between 10 and 20 ppb. The springtime values estimated for Los Angeles and Sacramento are less than 30 ppb. These concentrations are much lower than the actual springtime values observed at Trinidad Head. As indicated in Fig. 15, the GEOS-Chem model predictions for the spring (US EPA, 2012b) at Trinidad Head indicated that daily maximum 8-h average O₃ concentrations were consistently underestimated. Because Trinidad Head concentrations during the springtime are indicative of O₃ amounts reaching the California coast from the Pacific and these levels are underestimated by the GEOS-Chem model, it is more than likely that background levels for Los Angeles and Sacramento for the springtime

have also been underestimated. Additional support for this conclusion comes from others research results (Langford et al., 2011; Cooper et al., 2011; Lefohn et al., 2012-submitted; Lin et al., 2012-submitted).

5. Conclusion

In the first draft of the REA, the EPA believes that in estimating the risk from exposure to O₃ it is not necessary to take into consideration U.S. background concentrations beyond application of the quadratic rollback method. The current REA notes that the air quality assessment does not include estimates of background O₃, with the exception of providing a floor for O₃ concentrations when implementing the quadratic rollback method to simulate attainment of the current standards. Throughout the first draft of the REA, the EPA implies that U.S. background plays an unimportant role in the risk analysis.

For the second draft of the REA, the EPA Agency is assuming that estimates of background can be obtained directly from modeling. While acknowledging that the quadratic rollback method has its limitations, ignoring the limitations associated with the HDDM methodology may lead to great uncertainty in the estimation of U.S. background. If the model does not contain modules that can adequately address natural processes that contribute to background, such as stratospheric-tropospheric exchange, estimates of U.S. background levels will likely be subject to great uncertainties, which in turn, can affect risk estimates.

The uncertainty associated with the estimates of background O₃ has a major role in developing confidence in the risk assessment results described in the REA in Chapter 6 and Chapter 7. The level of estimated U.S. background is also important when assessing the reliability of the estimates of the Lowest Measured Level (LML). The "Heat Map" Tables

illustrate the estimated distribution of O₃-related all-cause mortality across distributions of daily 8-h maximum O₃ levels for each urban study area. In the REA, we note that 9 of the 12 cities are predicted to experience approximately 50% or more of O₃-related all-cause mortality for daily 8-h maximum O₃ levels between 25 and 50 ppb. For New York City, the EPA estimates that for recent conditions that 8-h daily maximum concentrations between 25 to 50 ppb result in 358 deaths or 54% of the estimated total all-cause mortality. Background O₃ concentrations are in the range of 25 to 50 ppb and at times, higher.

Background O₃ concentrations \geq 50 ppb frequently occur at Trinidad Head. Meteorological evidence exists to support the observation that conditions representative of U.S. background are routinely encountered at the low-elevation monitoring site at Trinidad Head, California. Long-range transport outside of North America and natural processes, such as stratospheric enhancement contribute to O₃ concentrations measured at this site. The frequency of hourly average concentrations \geq 50 ppb in the springtime, when almost all of the high concentrations occur, is large and varies from year to year during the month of April (e.g., 30 to 187 hours). In the period 2003-2011, daily maximum 8-h average O₃ concentrations sometimes exceeded 60 ppb during the springtime.

The range of U.S. background level concentrations is also important when characterizing health risks based on clinical studies. The REA illustrates the probabilistic exposure-response relationships for FEV₁ decrement \geq 10%. Although the probability of a response below 50 ppb is low, a large fraction of the population is estimated to be exposed to these low O₃ concentrations. These low O₃ concentrations are used in the risk model to predict the number of persons responding. Should the range of O₃ concentrations associated with U.S. background actually be much greater than predicted by the CMAQ model and more in line with the values observed at

Trinidad Head, then the risk predicted using clinical study results for the large fraction of the population may not be able to be reduced if reductions in emissions were to occur.

GEOS-Chem and other global models have difficulty representing the fine structures of O₃ events observed at relatively remote monitoring sites in the U.S., including events for which the contribution of U.S. background O₃ is likely important. Stratosphere-troposphere exchange can contribute to background O₃ at both low- and high-altitude sites. In addition, fire plumes transported on intercontinental scales can contain very high O₃ concentrations. In the ISA, the EPA notes that currently all transport models and not just the ones evaluated in the ISA, have difficulty in predicting day-specific quantities. Thus, estimates of U.S. background O₃ concentrations over a short-time frame (e.g., hourly or 8-h average concentrations) may not be reliable on a daily basis.

Results published in the literature indicate that the frequency of enhancements associated with stratospheric-tropospheric exchange processes can contribute to the variability of natural background. At many high- and low-elevation monitoring sites, numerous days occur in which a strong relationship exists between enhanced O₃ concentrations and stratosphere-to-troposphere transport that reaches the surface (STT-S) particularly during the springtime. When statistically significant coincidences occur at the lower elevation sites, the daily maximum hourly average concentrations are mostly in the 50-65 ppb range; the daily maximum 8-h average concentrations are usually in the 50-62 ppb range. Investigation of specific monitoring sites during periods when they were least affected by days with lightning, long-range transport from Asia, wildfires, or anthropogenic perturbations showed that high-elevation sites experience a greater percentage of days with concentrations ≥ 50 ppb and ≥ 60 ppb on days when STT-S >0 during the springtime. For example, for those days in which the STT-S >0 , the high-elevation Yellowstone

NP (WY) site experienced approximately 86% and 72% of the total days with 1-h and 8-h daily maximum concentrations ≥ 50 ppb, respectively. In comparison, the low-elevation sites at Voyageurs NP (MN), Cuyahoga County (OH), and Chittenden County (VT) experience lower percentages in the spring than the higher-elevation locations when STT-S >0 . Voyageurs NP experienced approximately 66% and 44% of the days with 1-h and 8-h daily maximum concentrations ≥ 50 ppb, respectively, while Cuyahoga County experienced approximately 64% and 45% of the days with 1-h and 8-h daily maximum concentrations ≥ 50 ppb, respectively. Chittenden County had approximately 66% and 51% of the days with 1-h and 8-h daily maximum concentrations ≥ 50 ppb, respectively.

The EPA illustrates in the REA the diurnal profiles of seasonally averaged hourly U.S. background floor values estimated for Los Angeles and Sacramento. The springtime values estimated for Los Angeles and Sacramento are less than 30 ppb. These concentrations are much lower than the actual springtime values observed at Trinidad Head. The ISA points out that the GEOS-Chem model predictions for the spring at Trinidad Head indicate that daily maximum 8-h average O₃ concentrations are consistently underestimated. Because Trinidad Head concentrations during the springtime are indicative of O₃ amounts reaching the California coast from the Pacific and these levels are underestimated by the GEOS-Chem model, it is more than likely that background levels for Los Angeles and Sacramento for the springtime have also been underestimated.

6. References

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. *Meteorol. Atmos. Phys.* 109, 9-18.

- Ambrose, J.L., Reidmiller, D.R., Jaffe, D.A., 2011. Causes of high O₃ in the lower free troposphere over the Pacific Northwest as observed at the Mt. Bachelor Observatory. *Atmos. Environ.* 45, 5302-5315.
- Bertschi, I.T., Jaffe, D.A., Jaegle, L., Price, H.U., Dennison, J.B., 2004. PHOBEA/ITCT 2002 airborne observations of transpacific transport of ozone, CO, volatile organic compounds, and aerosols to the northeast Pacific: Impacts of Asian anthropogenic and Siberian boreal fire emissions. *J. Geophys. Res.* 109, D23S12.
- Bertschi, I.T., Jaffe, D.A., 2005. Long-range transport of ozone, carbon monoxide, and aerosols to the NE Pacific troposphere during the summer of 2003: Observations of smoke plumes from Asian boreal fires. *J. Geophys. Res.* 110, D05303.
- Cooper, O., C. Forster, D. Parrish, E. Dunlea, G. Hübler, F. Fehsenfeld, J. Holloway, S. Oltmans, B. Johnson, A. Wimmers, L. Horowitz, 2004. On the life cycle of a stratospheric intrusion and its dispersion into polluted warm conveyor belts, *J. Geophys. Res.* 109, D23S09, doi:10.1029/2003JD004006.
- 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. *J. Geophys. Res.* 110, D23310, doi:10.1029/2005JD005783.
- Cooper, O.R., A. Stohl, M. Trainer, A.M. Thompson, J.C. Witte, S.J. Oltmans, G. Morris, K.E. Pickering, J.H. Crawford, G. Chen, R.C. Cohen, T.H. Bertram, P. Woolridge, A. Perring, W.H. Brune, J. Merrill, J.L. Moody, D. Tarasick, P. Nedelec, G. Forbes, M.J. Newchurch, F.J. Schmidlin, B.J. Johnson, S. Turquety, S.L. Baughcum, X. Ren, F.C. Fehsenfeld, J. Meagher, N. Spichtinger, C.C. Brown, S.A. Mckeen, I.S. McDermid, T. Leblanc, 2006. Large upper tropospheric ozone enhancements above mid-latitude North America during summer: In situ evidence from the IONS and MOZAIC ozone monitoring network, *J. Geophys. Res.*, 111, doi:10.1029/2006JD007306.
- Cooper, O.R., M. Trainer, A.M. Thompson, S.J. Oltmans, D.W. Tarasick, J.C. Witte, A. Stohl, S. Eckhardt, J. Lelieveld, M.J. Newchurch, B.J. Johnson, R.W. Portmann, L. Kalnajs, M.K. Dubey, T. Leblanc, I.S. McDermid, G. Forbes, D. Wolfe, T. Carey-Smith, G.A. Morris, B. Lefer, B. Rappenglück, E. Joseph, F. Schmidlin, J. Meagher, F.C. Fehsenfeld, T. J. Keating, R.A. Van Curen, K. Minschwaner, 2007. Evidence for a recurring eastern North America ozone maximum during summer, *J. Geophys. Res.* 112, doi:10.1029/2007JD008710.
- Cooper, O.R., Oltmans, S.J., Johnson, B.J., Brioude, J., Angevine, W., Trainer, M., Parrish, D.D., Ryerson, T.R., Pollack, I., Cullis, P.D., Ives, M.A., Tarasick, D.W., Al-Saadi, J., Stajner, I., 2011. Measurement of western U.S. baseline ozone from the surface to the tropopause and assessment of downwind impact regions. *J. Geophys. Res.* 116, D00V03, doi:10.1029/2011JD016095 doi:10.1029/2011JD016095.

- Cristofanelli, P., Bonasoni, P., Tositti, L., Bonafe', 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). *J. Geophys. Res.* 111, D03306, doi:10.1029/2005JD006553.
- Danielsen, E.F., 1968. Stratospheric-tropospheric exchange based on radioactivity, ozone and potential vorticity. *J. Atmos. Sci.* 25, 502-518.
- 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. *Atmos. Environ.* 28, 53-68.
- 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. *Atmos. Environ.* 47, 206-217.
- 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. *J. Geophys. Res.* 108 (D24), 4787, doi:10.1029/2003JD003855.
- Goldstein, A.H., Millet, D.B., McKay, M., Jaegle, L., Horowitz, L., Cooper, O., Hudman, R., Jacob, D.J., Oltmans, S., Clarke, A., 2004. Impact of Asian emissions on observations at Trinidad Head, California during ITCT 2K2, *J. Geophys. Res.* 109, D23S17, doi:10.1029/2003JD004406.
- 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.
- 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 windprofiler radars. *Nature* 450, 281-284.
- Huang, M., G.R. Carmichael, B. Adhikary, S.N. Spak, S. Kulkarni, Y. Cheng, C. Wei, Y. Tang, D.D. Parrish, S.J. Oltmans, A. D'Allura, A. Kaduwela, C. Cai, A.J. Weinheimer, M. Wong, R.B. Pierce, J.A. Al-Saadi, D.G. Streets, Q. Zhang, 2010. Impacts of transported background ozone on California air quality during the ARCTAS-CARB period; a multi-scale modeling study, *Atmos. Chem. Phys.* 10, doi:10.5194/acp-10-6947-2010, 6947-6968.
- Jaffe, D., Bertschi, I., Jaegle, L., Novelli, P., Reid, J.S., Tanimoto, H., Vingarzan, R., Westphal, D.L., 2004. Long-range transport of Siberian biomass burning emissions and impact on surface ozone in western North America *Geophys. Res. Lett.* 31, 16106, doi:10.1029/2004GL020093.

- Langford, A.O., Aikin, K.C., Eubank, C.S., Williams, E.J., 2009. Stratospheric contribution to high surface ozone in Colorado during springtime. *Geophys. Res. Lett.* 36, L12801, doi:10.1029/2009GL038367.
- Langford, A.O., Brioude, J., Cooper, O.R., Senff, C.J., Alvarez II, R.J., Hardesty, R.M., Johnson, B.J., Oltmans, S.J., 2011. Stratospheric influence on surface ozone in the Los Angeles area during late spring and early summer of 2010. *J. Geophys. Res.* 117, D00V06, doi:10.1029/2011JD016766.
- Lefohn, A.S., Shadwick, D.S., and Ziman, S.D., 1998. The difficult challenge of attaining EPA's new ozone standard, *Environ. Sci. and Tech.* 32, 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.
- 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. *Atmos. Environ.* 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. Manuscript submitted for publication.
- Lin, M., Fiore, A.M., Horowitz, L.W., Cooper, O.R., Naik, V., Holloway, J., Johnson, B.J., Middlebrook, A.M., Oltmans, S.J., Pollack, I.B., Ryerson, T.B., Warner, J.X., Wiedinmyer, C., Wilson, J., Wyman, B., 2012. Transport of Asian ozone pollution into surface air over the western United States in spring. *J. Geophys. Res.* 117, D00V07, doi:10.1029/2011JD016961.
- Lin, M., Fiore, A.M., Cooper, O.R., Horowitz, L.W., Langford, A.O., Levy II, H., Johnson, B.J., Vaishali, N., Oltmans, S.J., Senff, C.J., 2012. Springtime high surface ozone events over the western United States: Quantifying the role of stratospheric intrusions, *J. Geophys. Res.* Manuscript submitted for publication.
- 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.
- 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. *Environ. Sci. & Tech.* 45, doi:10.1021/es2022918, 9484-9497.

- Oltmans, S.J., Lefohn, A.S., Harris, J.M., Shadwick, D., 2008. Background ozone levels of air entering the west coast of the U.S. and assessment of longer-term changes. *Atmos. Environ.* 42, 6020-6038.
- Oltmans, S.J., Lefohn, A.S., Harris, J.M., Tarasick, D.W., Thompson, A.M., 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. *Atmos. Environ.* 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. *Geophys. Res. Lett.* 34, L07805, doi:10.1029/2006GL029113.
- Parrish, D.D., D.B. Millet, A.H. Goldstein, 2009. Increasing ozone in marine boundary layer inflow at the west coasts of North America and Europe, *Atmos. Chem. Phys.* 9, 1303-1323.
- Parrish, D.D., Aikin, K.C., Oltmans, S.J., Johnson, B.J., Ives, M., Sweeny, C., 2010. Impact of transported background ozone inflow on summertime air quality in a California ozone exceedance area. *Atmos. Chem. Phys.* 10, 10093-10109, doi:10.5194/acp-10-10093.
- Pfister, G.G., Emmons, L.K., Edwards, D.P., Arellano, A., Sachse, G., Campos, T., 2010. Variability of springtime transpacific pollution transport during 2000-2006: the INTEX-B mission in the context of previous years. *Atmos. Chem. Phys.* 10, 1345-1359.
- Reed, R.J., 1955. A study of a characteristic type of upper level frontogenesis. *J. Meteorol.* 12, 226-237.
- 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. *Atmos. Environ.* 33, 1735-1744.
- 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, *Atmos. Environ.* 34, 1323-1354.
- US Environmental Protection Agency, US EPA, 1996. Air Quality Criteria for Ozone and Related Photochemical Oxidants. EPA/600/P-93/004af. Research Triangle Park, NC: U.S. Environmental Protection Agency, Office of Research and Development.
- US Environmental Protection Agency, US EPA, 2009. Ozone design values for 2008-2010. <http://www.epa.gov/airtrends/values.html>.

- US Environmental Protection Agency, US EPA, 2012a. Health Risk and Exposure Assessment for Ozone. EPA/452/P-12-001. Research Triangle Park, NC: Office of Air Quality Planning and Standards. July.
- US Environmental Protection Agency, US EPA, 2012b. Integrated Science Assessment for Ozone and Related Photochemical Oxidants. EPA/600/R-10/076C. Research Triangle Park, NC: Office of Research and Development. June.
- Weiss-Penzias, P., Jaffe, D.A., Swartzendruber, P., Dennison, J.B., Chand, D., Hafner, W., Prestbo, E., 2006. Observations of Asian air pollution in the free troposphere at Mount Bachelor Observatory during the spring of 2004. *J. Geophys. Res.* 111 (D10304). doi:10.1029/2005JD006522.
- 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.
- 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. *Atmos. Environ.* 45, 6769-6776.

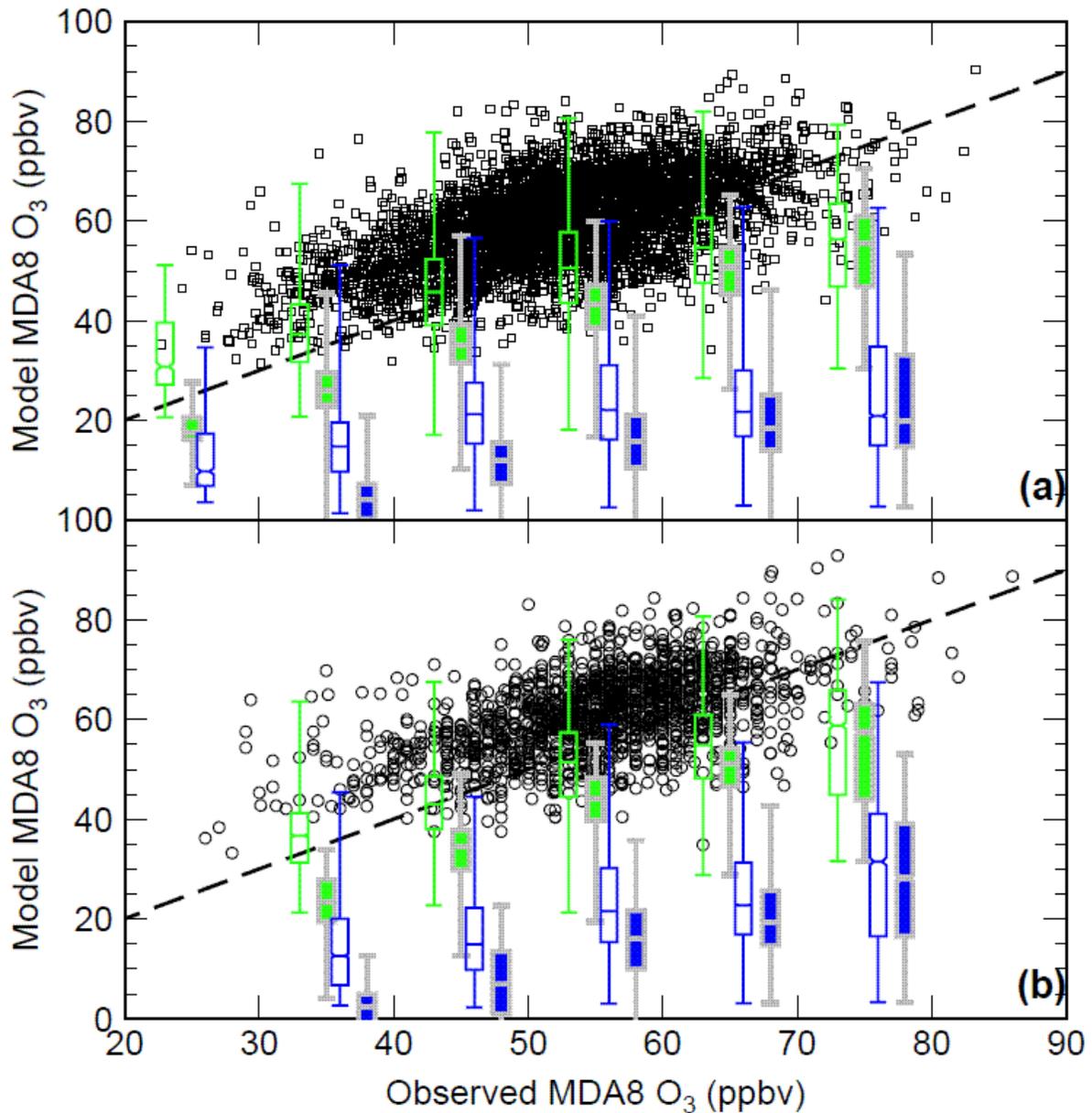


Fig. 1. Observed versus simulated MDA8 surface O₃ for April-June 2010 at (a) AQS sites over the central western U.S. and (b) 15 high-elevation sites. Also shown is the 1:1 line. The box-and-whisker plots (minimum, 25th, 50th, 75th percentiles, and maximum) give statistics of the NA background (green) and the stratospheric contribution (blue) for every 10-ppb bin of observed values. Points greater than 80 ppbv are merged to the 70-80 ppbv range. The filled boxes represent the bias-corrected estimates by assuming that model overestimates of total O₃ are entirely driven by excessive stratospheric influence. Source: Lin et al. (2012-submitted).

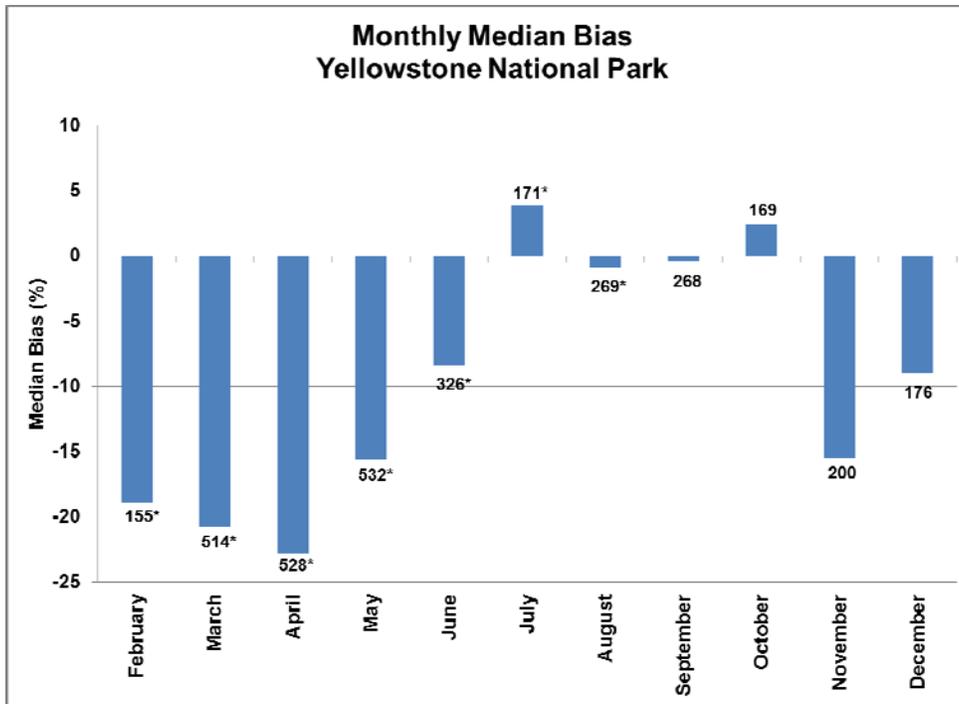


Fig. 2. Monthly median bias between modeled and observed hourly O₃ at Yellowstone NP for 2006. The total STT-S counts for each month are indicated; a statistically significant month is indicated with an asterisk (*).

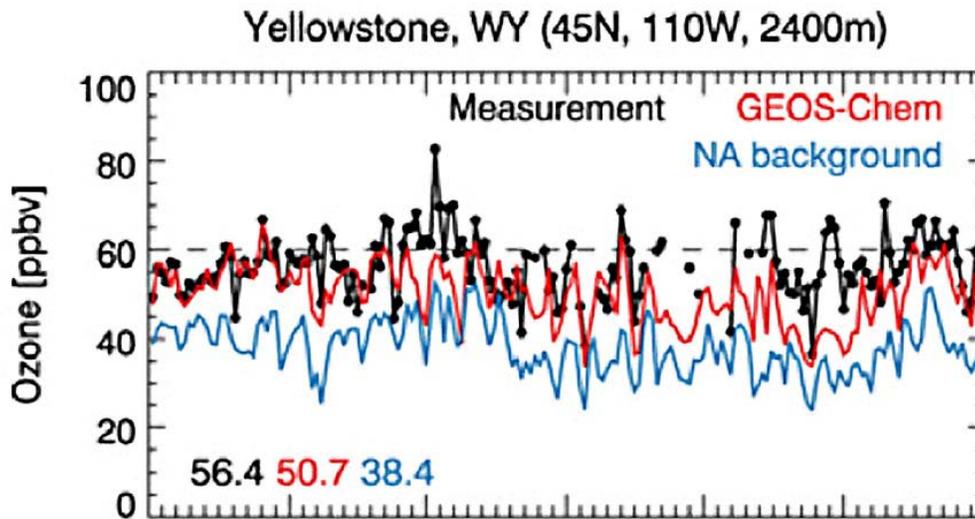


Fig. 3. Time series of measurements of daily maximum 8-h average ozone concentrations at Yellowstone NP with GEOS-Chem predictions for the base case and for the North American background case during March-August, 2006. Source: EPA (2012b).

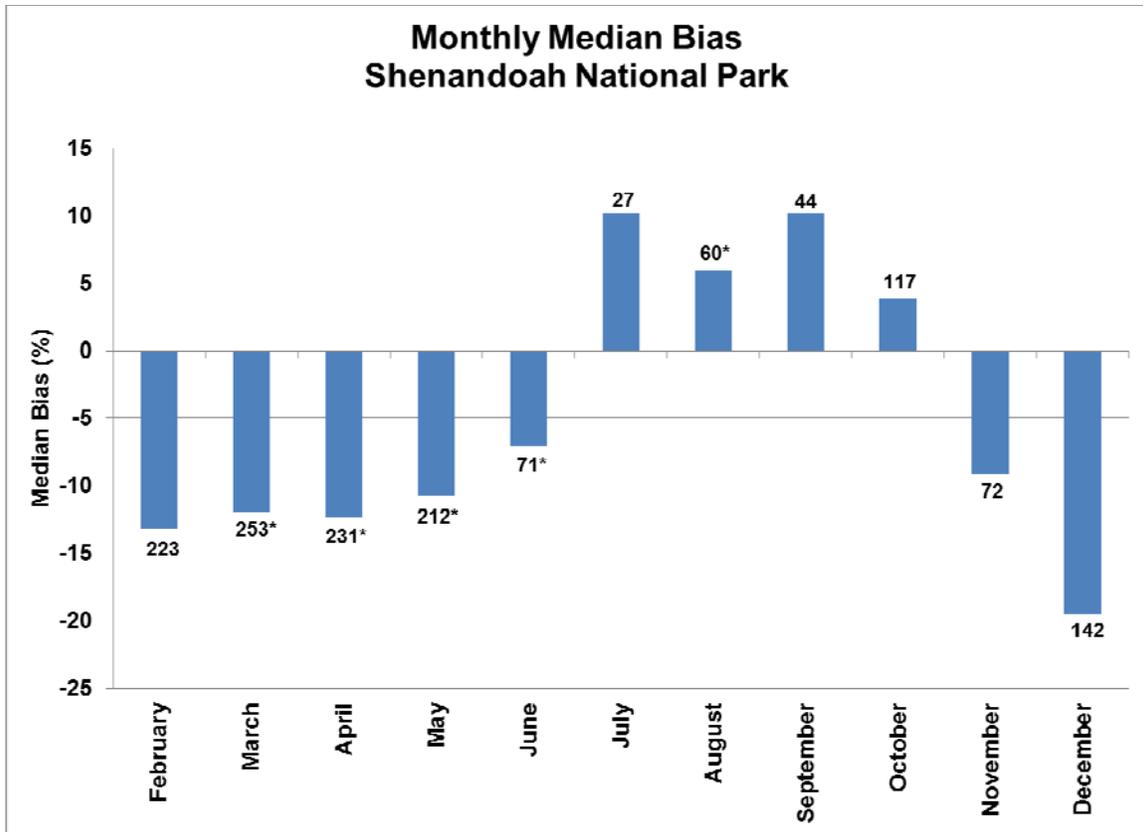


Fig. 4. Monthly median bias between modeled and observed hourly O₃ at Shenandoah National Park for 2006. The total STT-S counts for each month are indicated; a statistically significant month is indicated with an asterisk (*).

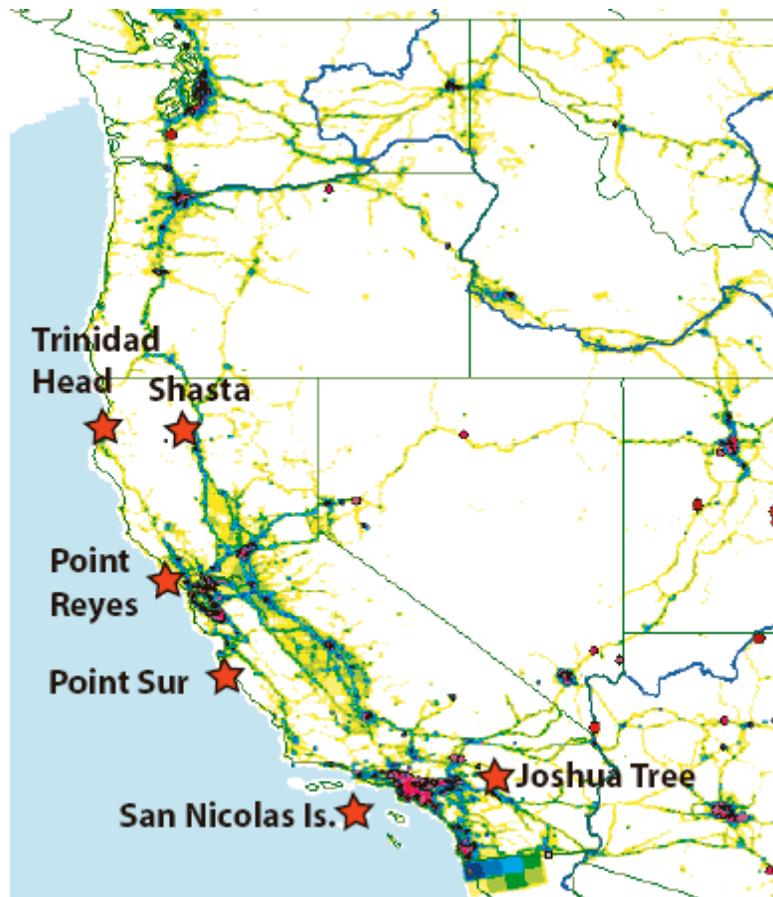


Fig. 5. Map of the location of the surface O₃ and ozonesonde measurement site at Trinidad Head and ozonesonde sites in California during the IONS 2010/CalNex campaign.

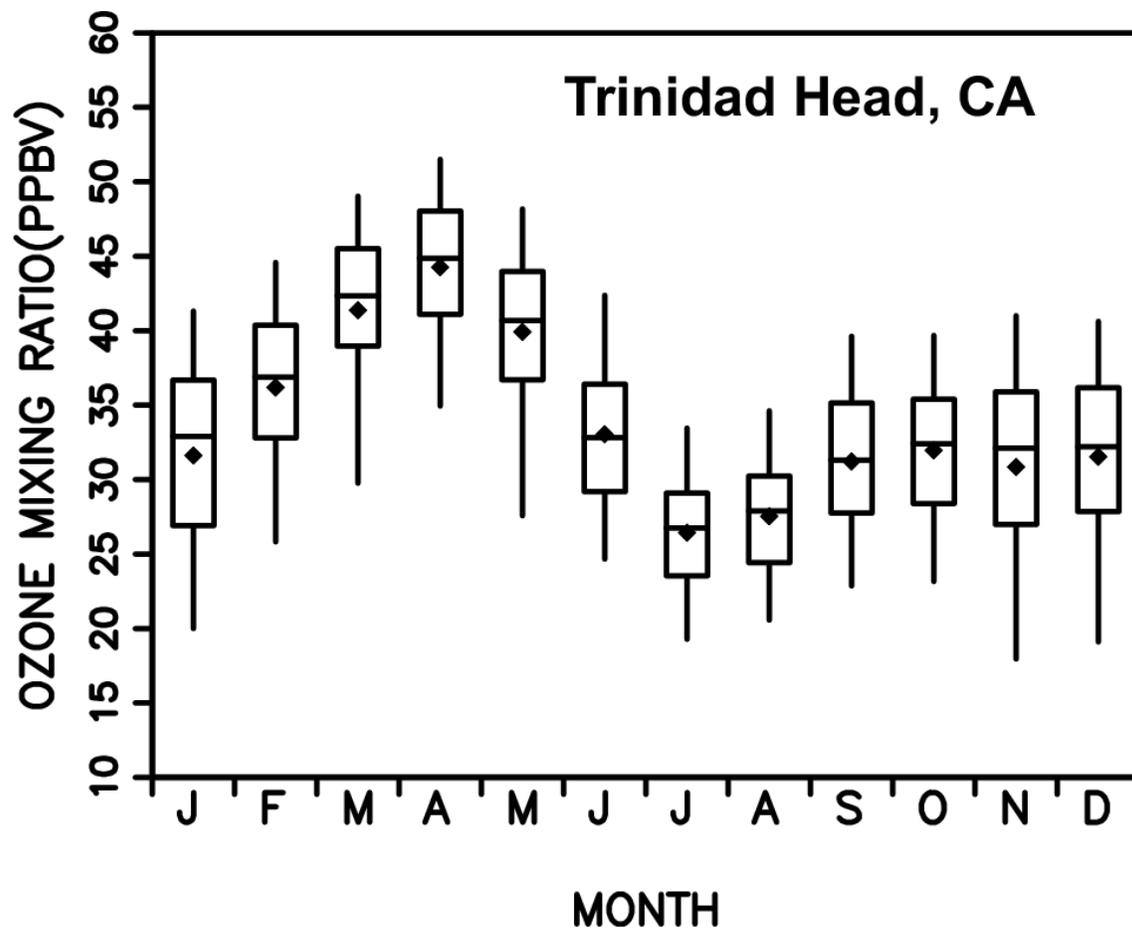


Fig. 6. Seasonal variation of daytime (1100-1800 Local Standard Time) surface O₃ at Trinidad Head, California (41N). The diamond is the mean, the horizontal bar within the box is the median, the box is the inner 50th percentile and the whiskers are the inner 90th percentile of the hourly averages based on data from 2002-2010.

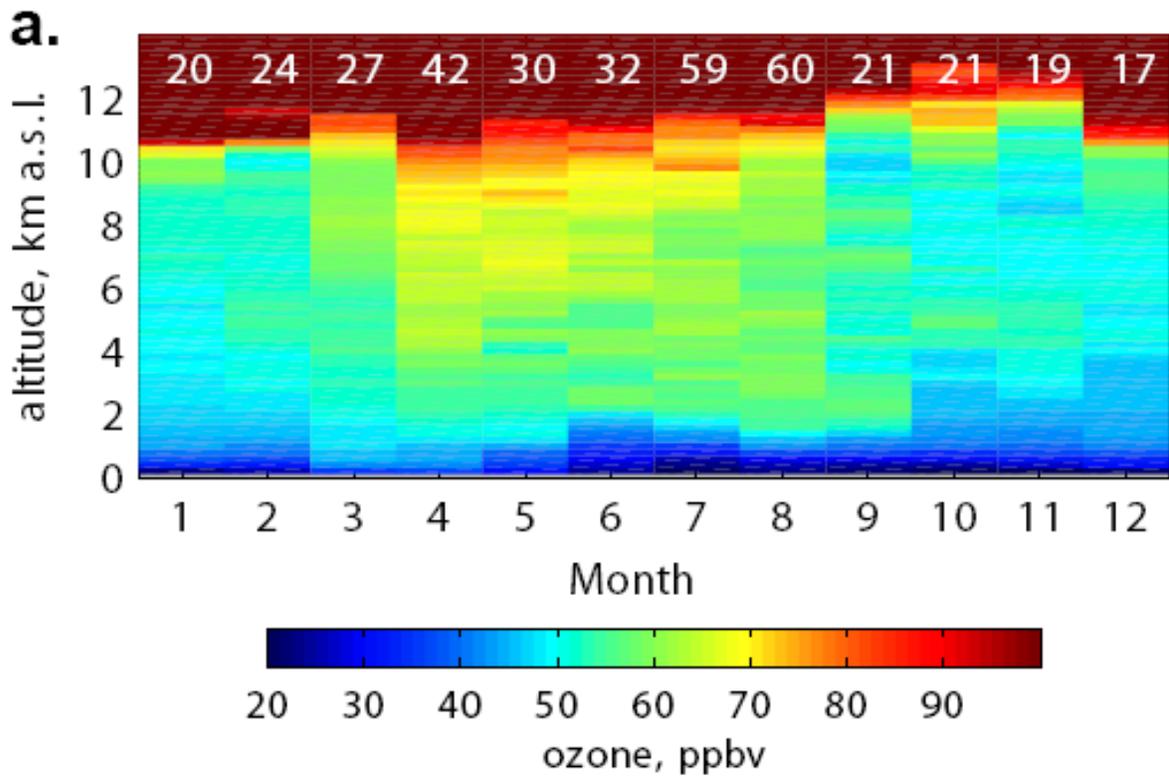


Fig.7. Tropospheric O₃ mixing ratios (ppbv) above the Trinidad Head Observatory from ozonsonde measurements. The numbers at the top of the figure for each month are the number of soundings for that month in the period 2004-2010. Source: Cooper et al. (2011).

Trinidad Head

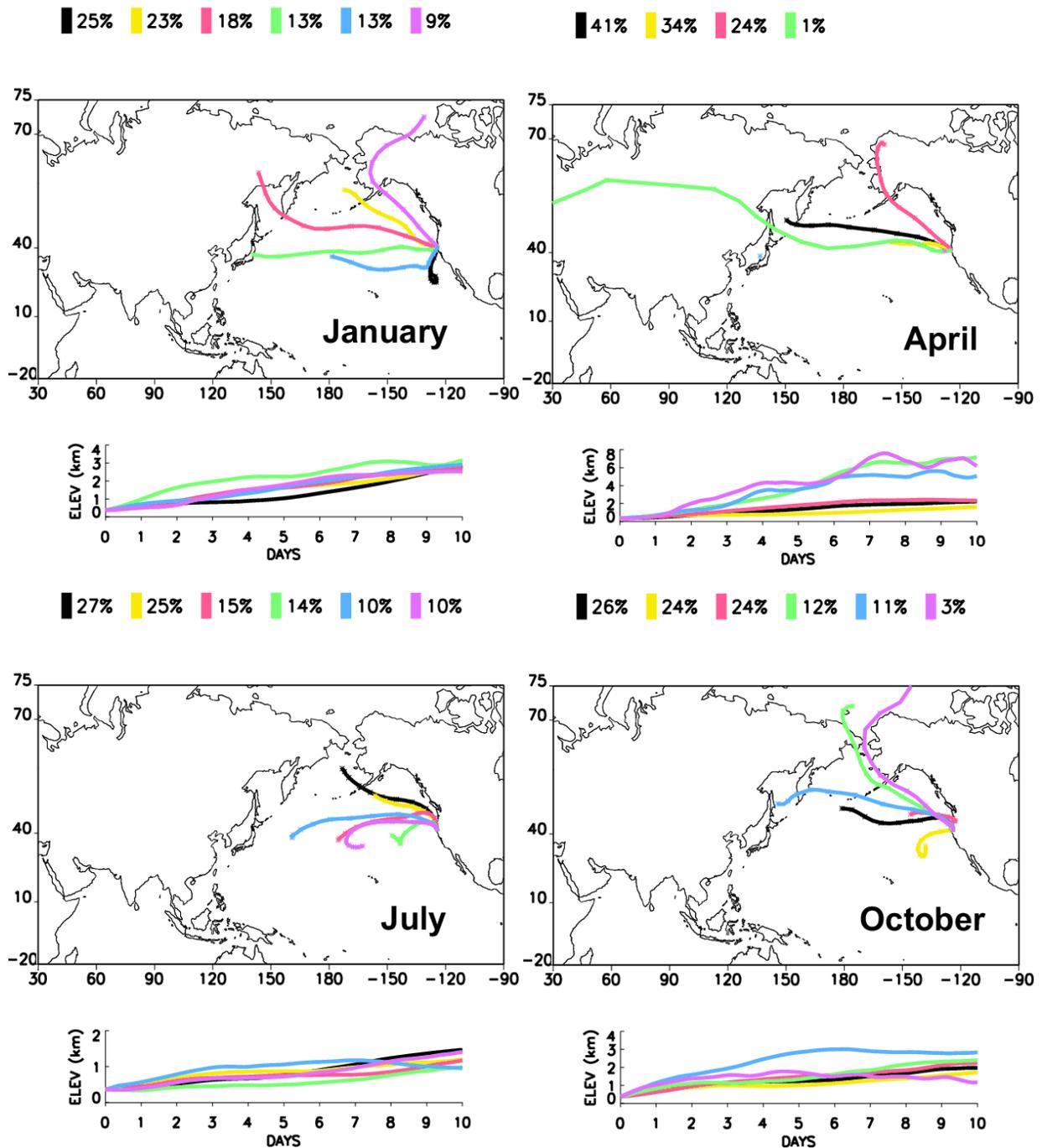


Fig. 8. Trajectory clusters for Trinidad Head, California for years 2002 to 2005 showing average flow characteristics at the station. Source: Oltmans et al. (2008).

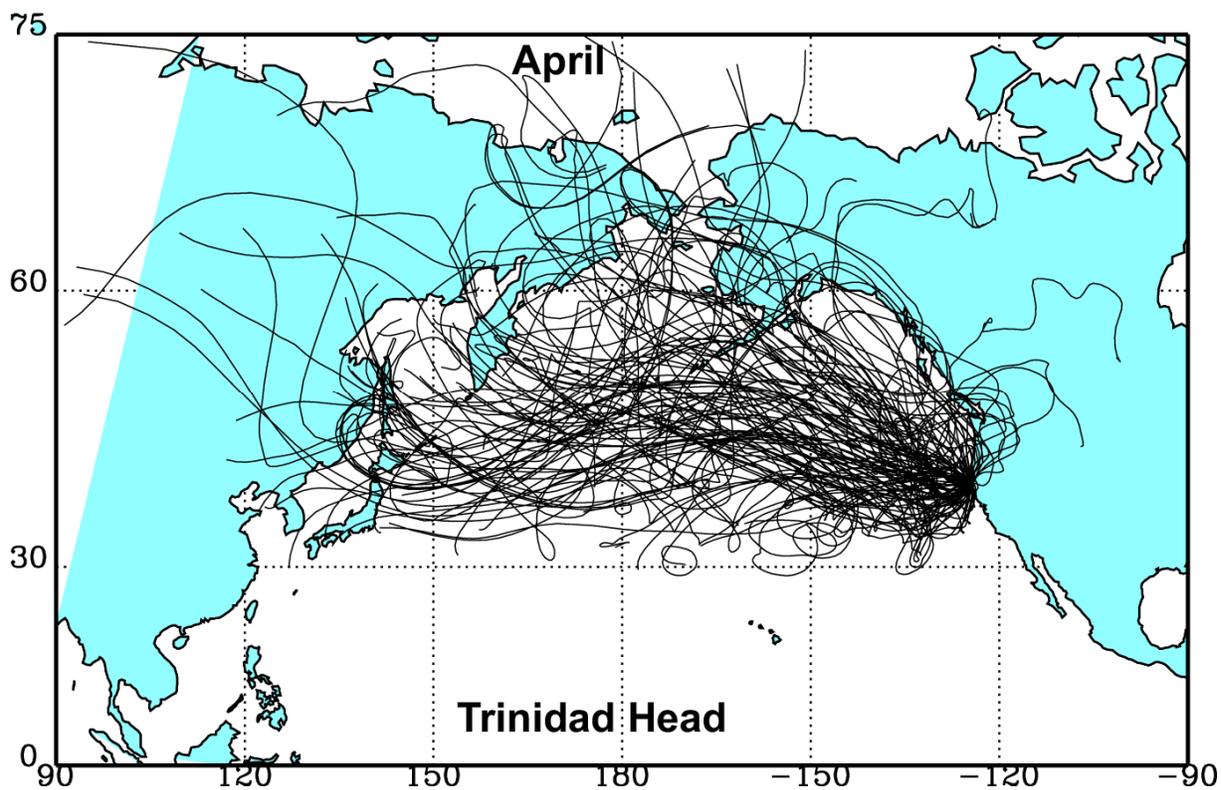


Fig. 9. Back trajectories from Trinidad Head, California on days with hourly average O_3 amounts ≥ 50 ppb for April in the years 2002 to 2005. Source: Oltmans et al. (2008).

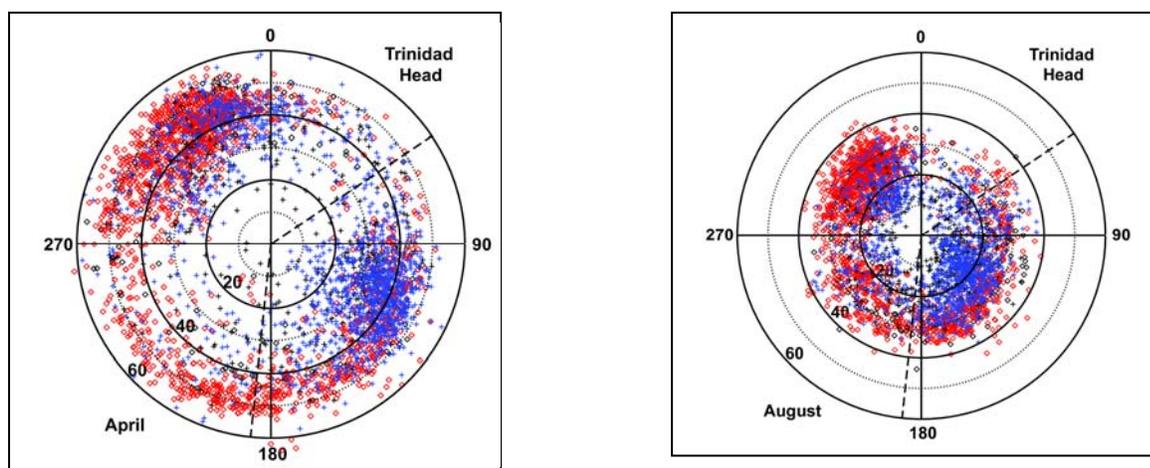


Fig. 10. Wind rose for O_3 mixing ratios at Trinidad Head for April and August for the period 2002-2007. The concentric circles mark the O_3 mixing from 0 ppbv at the center to 60 ppbv for the outer circle. The red diamonds are daytime (10-21 LST) values and the blue pluses are nighttime (22-09 LST) values. When wind speeds are less than 0.5 m/s the symbol is black. The wind direction is labeled by the direction from which the wind is blowing. Between the dashed lines (56° to 186°), the wind is blowing from over the land to the observing site. Source: Oltmans et al. (2008).

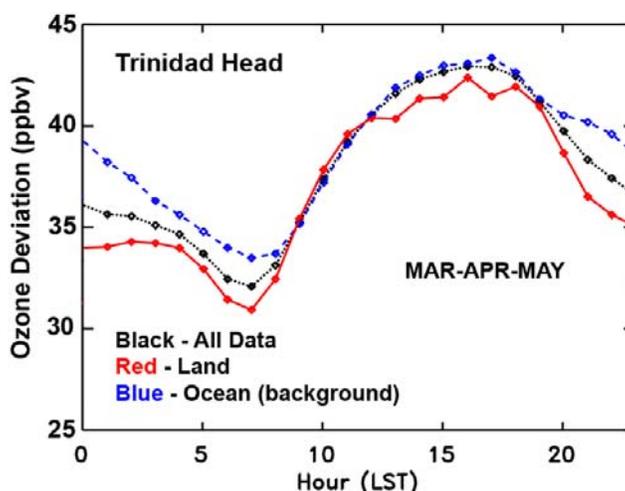


Fig. 11. Diurnal variation of O_3 mixing ratio at Trinidad Head for March-April-May for all observations (black), for observations when air reaching the observing site has recently come from over land (red), and for observations when air is coming from off the ocean (blue). Air reaching the site from off the ocean is overwhelmingly representative of background conditions. Source: Adapted from Oltmans et al. (2008).

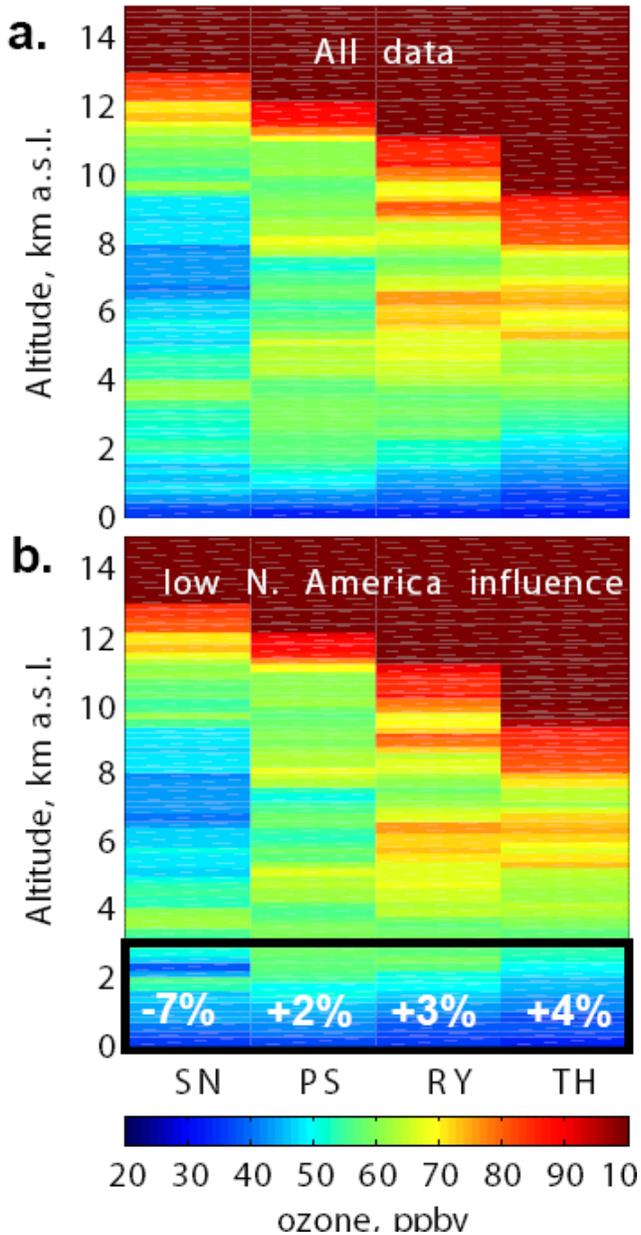


Fig. 12. a) Median O₃ profiles above the four coastal sites using all available data. b) Same as in a. but measurements with recent North American anthropogenic influence removed. Numbers in white indicate the percent change in the mass of O₃ in the 0-3 km range when air masses with strong North American influence are removed. None of these changes are statistically significant. Source: Cooper et al. (2011).

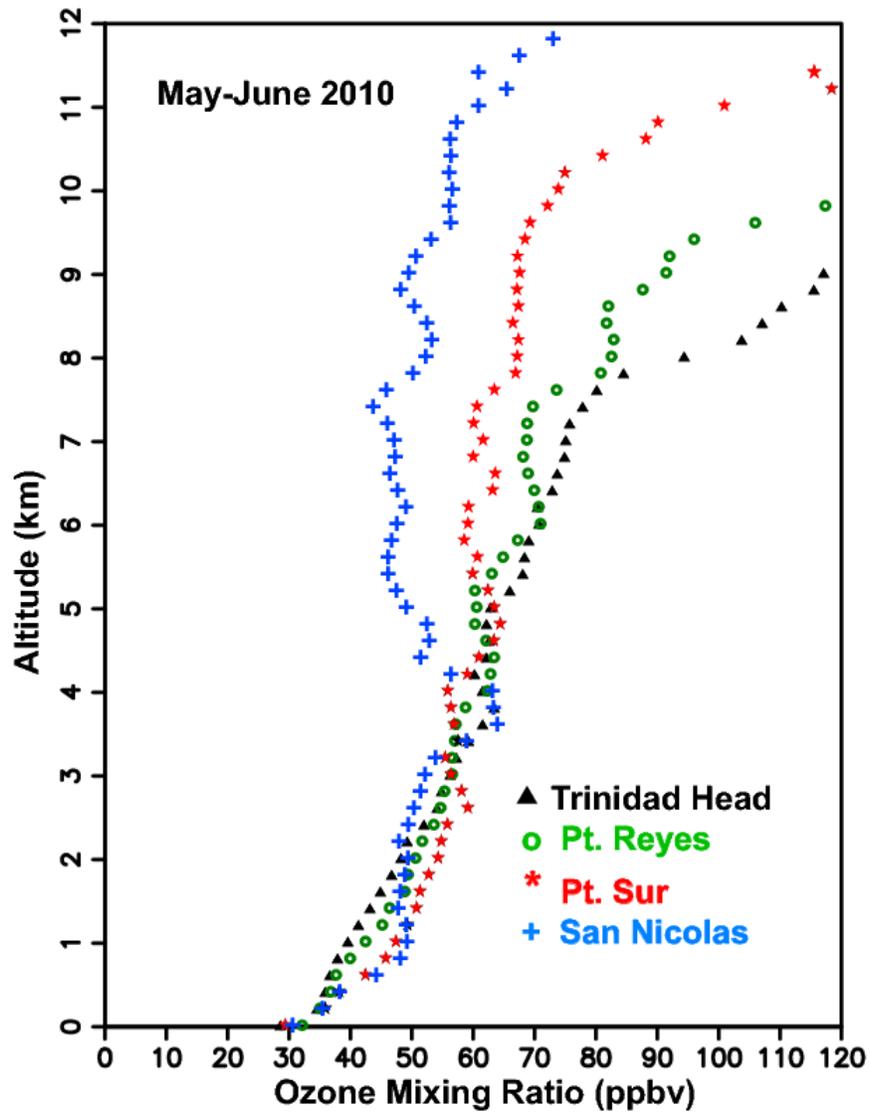


Fig. 13. Average O₃ mixing ratio profiles at four sites making ozonesonde observations during the IONS 2010/CalNex Campaign in May-June 2010. In the lower troposphere (<4 km) average O₃ amounts are similar along the California coast.

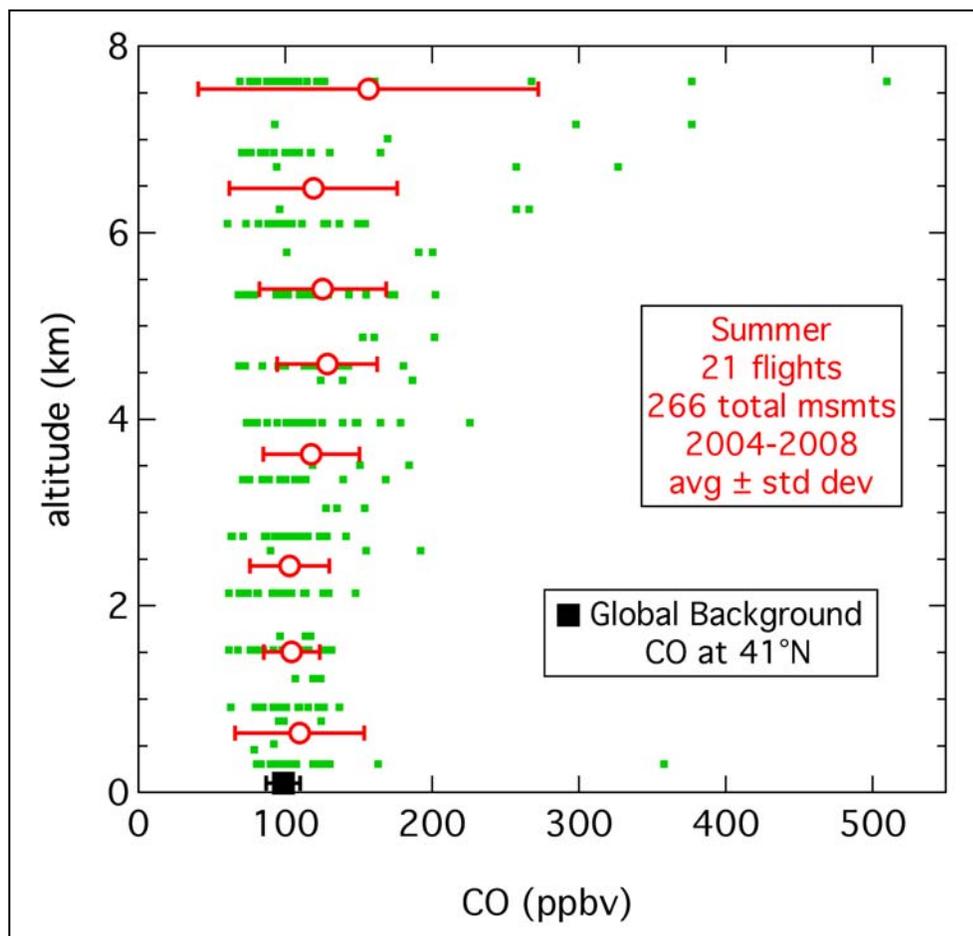
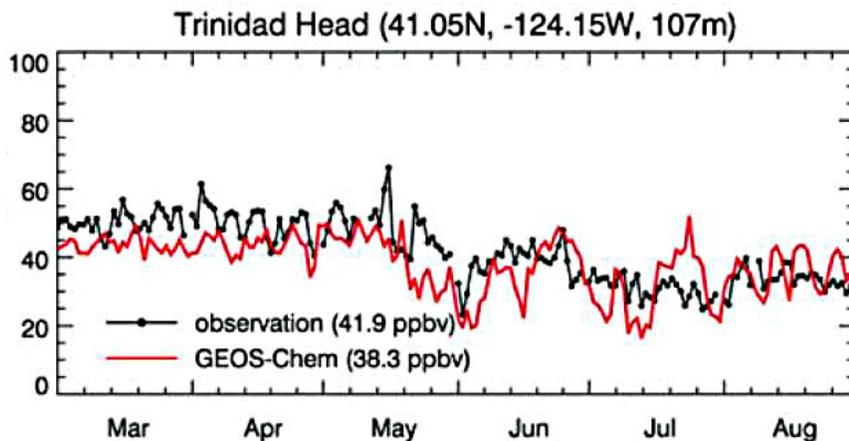


Fig. 14. Vertical profiles of carbon monoxide measured in flasks collected on aircraft flights above Trinidad Head CA. The green points give the individual measurements, and the red circles indicate averages and standard deviations for 1 km altitude segments. The black symbol gives the surface global carbon monoxide background as determined from the NOAA ESRL Global Monitoring Division flask network for the years of the aircraft measurements. Source: Parrish et al. (2010).

a)



b)

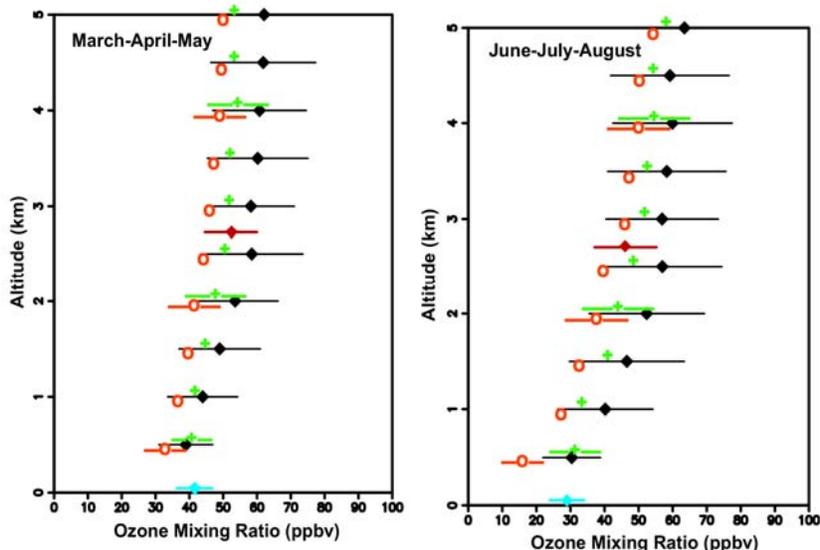


Fig. 15. a) Comparison of daily maximum 8-h average O_3 predicted using GEOS-Chem at $0.5^\circ \times 0.667^\circ$ with measurements at Trinidad Head, CA from March to August 2006. Source: US EPA (2012b). b) Average spring (March/April/May) O_3 profiles derived from ozone sondes at Trinidad Head are shown in the left panel for 1997 through 2010. These data represent averages over 500 meters in altitude with ± 1 standard deviation. The symbols in blue are the seasonal surface O_3 average at Trinidad Head and the symbol in red is the Mt. Bachelor seasonal average plotted at the altitude of the observatory. Profile results from the GEOS-Chem model for 2006 are shown as green pluses. The contribution from Policy Relevant Background in the model profile is shown by orange circles. Representative standard deviations at several altitudes for the model are shown. The right panel shows O_3 profiles during the summer months (June/July/August) at Trinidad Head and from the model. The model results are plotted slightly offset in altitude from the nominal altitude for clarity. Source: McDonald-Buller et al. (2011).

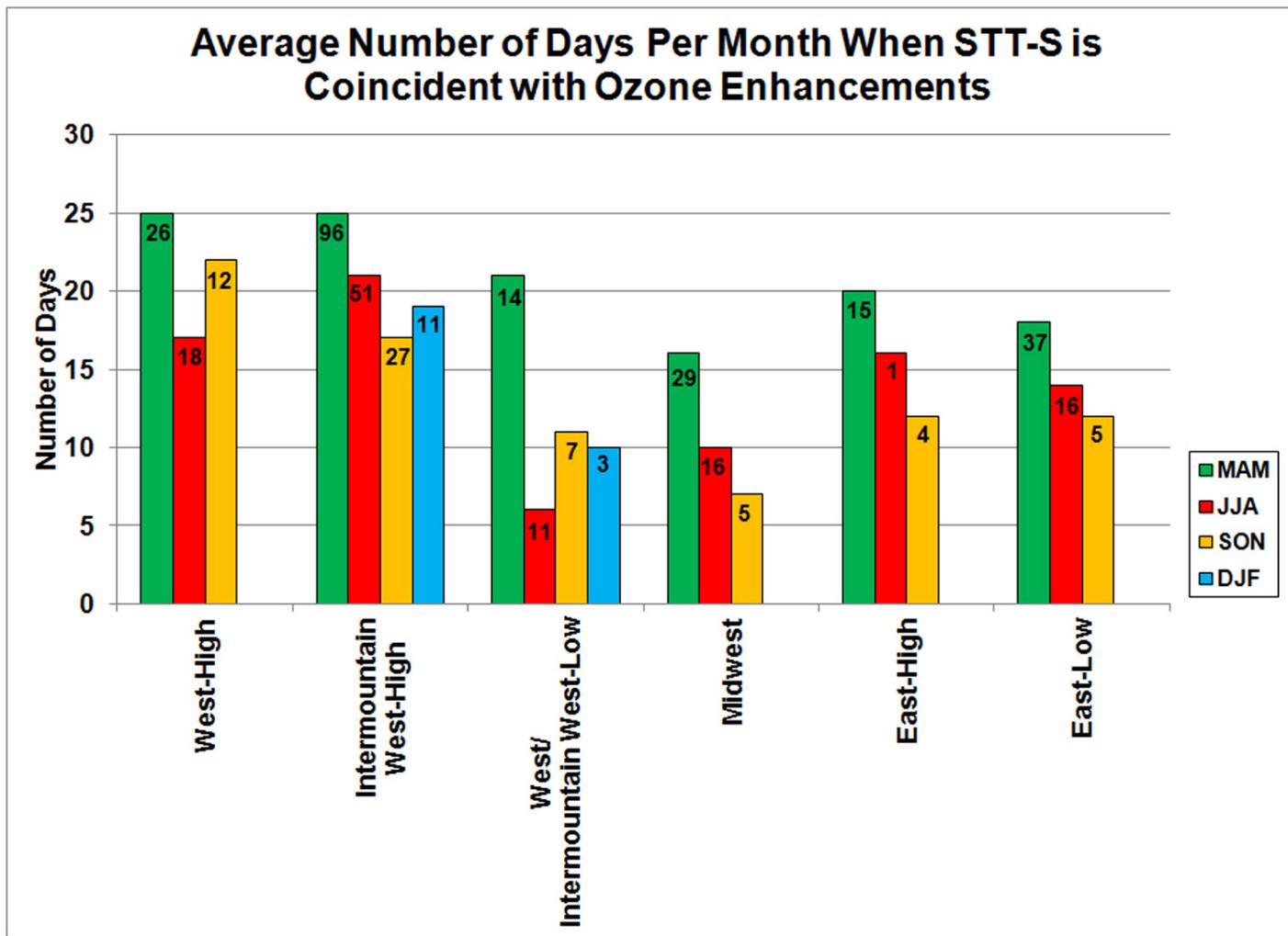


Fig. 17. Average number of days per month when STT-S is coincident with O₃ enhancements by geographic region for spring (March, April, and May), summer (June, July, and August), fall (September, October, and November), and winter (December, January, and February). The numbers within each bar are the number of site-months contributing to the average. Source: Lefohn et al. (2012-submitted)

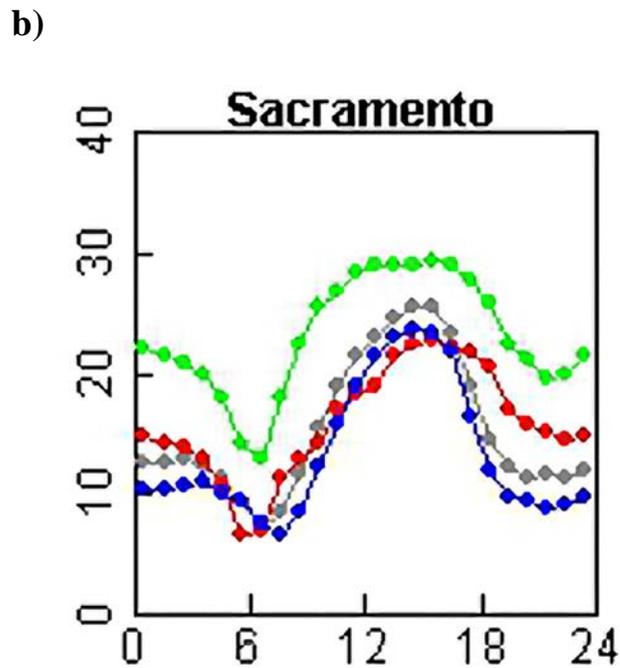
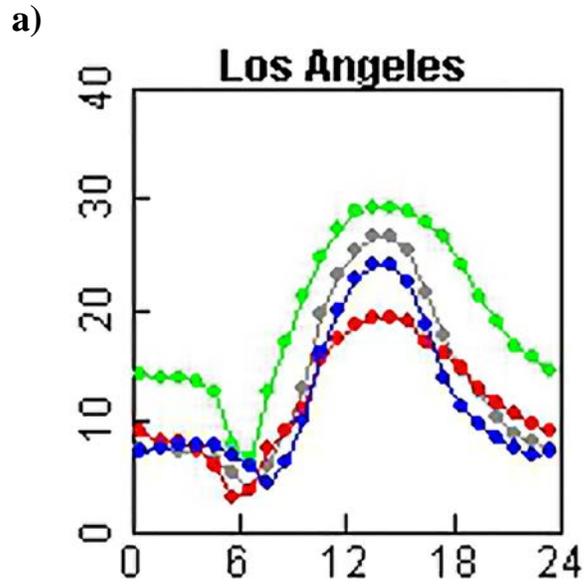


Fig. 18. Diurnal profiles of seasonally averaged U.S. background floor values estimated for a) Los Angeles and b) Sacramento. Profiles for spring, summer, autumn, and winter are colored in green, red, gray, and blue, respectively. Source: US EPA (2012b; page 4-9.)