

Background Ozone Concentrations

Comments Relating to the Third Draft of the Integrated Science Assessment for Ozone and Related Photochemical Oxidants

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 and Definition of Background

- The estimation of background O₃ concentrations is extremely important in order to adequately fulfill the Agency's goal of assessing the risk associated with O₃ exposures. Too low an estimate can result in (1) unreliable statistical significance outcomes associated with controlled laboratory exposure studies, (2) overestimated human health risk predictions, and (3) optimistic policy expectations of the levels to which hourly average O₃ concentrations can be lowered as a result of emission reduction requirements.
- The most recent draft of the Integrated Science Assessment (ISA) defines background concentrations of O₃ in various ways:

Policy Relevant Background (PRB) O₃ has been supplanted by North American background (NA background) O₃.

Natural background in the current version of the ISA “includes contributions resulting from emissions from natural sources (e.g., stratospheric intrusions, wildfires, biogenic methane and more short-lived VOC emissions) throughout the globe simulated in the absence of anthropogenic emissions”.

North American background “includes contributions from natural background throughout the globe and emissions from anthropogenic pollutants contributing to global concentrations of O₃ ... from countries outside of North America.

United States background “includes contributions from natural background throughout the globe and emissions from anthropogenic pollutants from countries

outside the U.S.” (i.e., it includes emissions from Canada and Mexico).

- 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.

ES-2. Comparing Model Results with Observational Data

- 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 modeling results were capable of replicating the 4th highest MDA8 O₃ to within suitable bounds on a day-specific basis. The EPA points out that currently all chemical transport models and not just the ones evaluated in the ISA, have difficulty in predicting day-specific quantities. The difficulty in replicating day specific base-case O₃ concentrations within reasonable bounds appears to be related to the models’ respective resolutions, uncertainties in their input data sets, and the specific modules used to describe specific processes (e.g., stratospheric-tropospheric exchange).
- 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) that are 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 the EPA uses in its risk assessment methodology 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 (US EPA, 2012a) 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). 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 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 (US EPA, 2012a) 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.

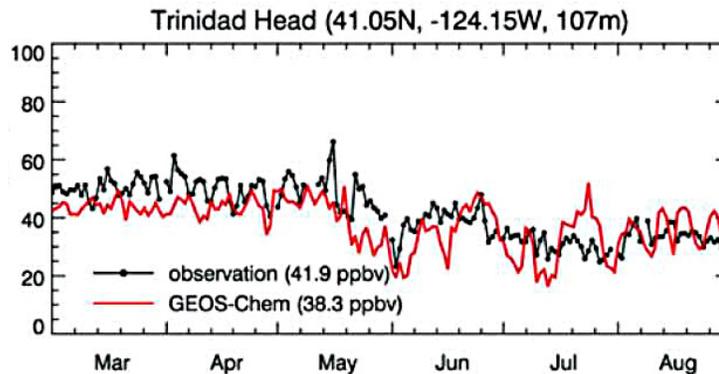


Fig. E-1. Comparison of daily maximum 8-h average O₃ predicted using GEOS-Chem at $0.5^\circ \times 0.667^\circ$ with measurements at Trinidad Head, CA from March to August 2006. Source: US EPA (2012a).

- 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. An example from the preliminary analysis is provided 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 STE contribution. STT counts in the figure 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.

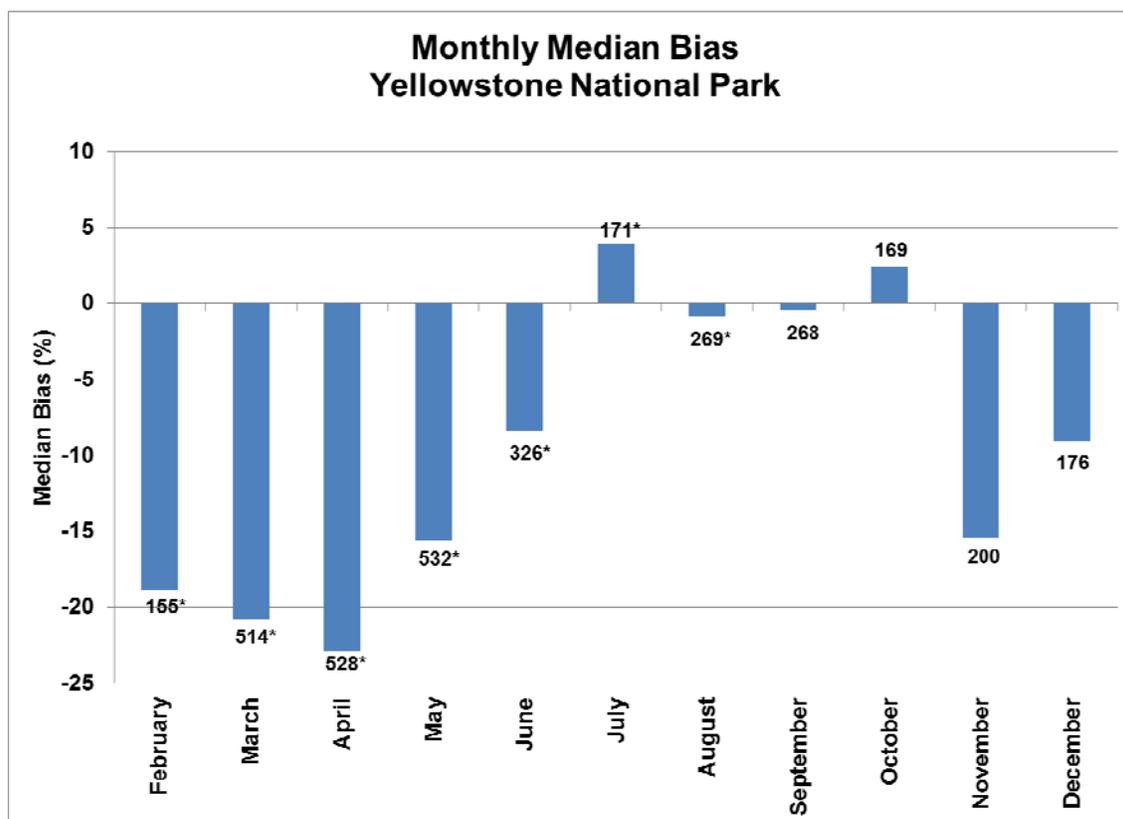


Fig. E-2. Monthly median bias between modeled and observed hourly O₃ at Yellowstone NP for 2006. The total STT-S counts for each month are indicated; a statistically significant month is indicated with an asterisk (*).

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

- 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.
- 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. Introduction

As noted in the most recent draft of the Integrated Science Assessment (ISA) dated June 2012 (US EPA, 2012a), background concentrations of O₃ have been defined in various ways. The earlier term, Policy Relevant Background or PRB O₃ (US EPA, 2006; 2011) was supplanted by North American background (NA background) O₃ (US EPA, 2011). The current draft also defines natural background that “includes contributions resulting from emissions from natural sources (e.g., stratospheric intrusions, wildfires, biogenic methane and more short-lived VOC emissions) throughout the globe simulated in the absence of anthropogenic emissions”. North American background “includes contributions from natural background throughout the globe and emissions from anthropogenic pollutants contributing to global concentrations of O₃ ... from countries outside of North America. The ISA notes that United States background “includes contributions from natural background throughout the globe and emissions from anthropogenic pollutants from countries outside the U.S.” (i.e., it includes emissions from Canada and Mexico).

As noted on page 7-16 of the first draft of the Health Risk and Exposure Assessment for Ozone document (US EPA, 2012b), the EPA no longer focuses on model-based PRB/NA background estimates but rather on model-based estimates of U.S. background. Estimated U.S. background O₃ level estimates are used in the risk analysis methodology as a lower bound for hourly O₃ concentrations in the quadratic rollback procedure to simulate just meeting the current standard level. However, 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, 2012b). In addition, the range of U.S. background level concentrations is important when characterizing health risks based on clinical studies. A large fraction of the population is estimated to be exposed to low O₃

concentrations (US EPA, 2012b). 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 using the GEOS-Chem 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. As will be discussed in our report, the Agency's assumption that hourly averaged U.S. background O₃ concentrations are generally very low across the entire US appears to be questionable.

To explore the uncertainty in the estimate of U.S. background hourly average O₃ concentrations, it is important to evaluate the limitations associated with the modeling estimates described in the ISA (US EPA, 2012a). As noted McDonald-Buller et al. (2011) and EPA (2012b), the EPA requires the use of the time series of hourly averaged O₃ concentrations to determine for the epidemiological risk analysis a range of exposure metrics, such as (a) daily maximum 1-hour concentrations, (b) daily maximum 8-h concentrations, (c) daytime 8-h average concentrations (10:00AM to 6:00PM), and (d) daily 24-h average concentrations.

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 and evaluated based on observational data. McDonald-Buller et al. (2011) note 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. The authors 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 EPA describes some of the modeling limitations noted in McDonald-Buller et al. (2011). On page 3-63 in the ISA (US EPA, 2012a), 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, 2012a), 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.

Characterizing U.S. background O₃ levels that are too low can result in (1) unreliable statistical significance outcomes associated with controlled laboratory exposure studies, (2) overestimated human health risk predictions, and (3) optimistic policy expectations of the levels to which hourly average O₃ concentrations can be lowered as a result of emission reduction requirements. Because of the importance of correctly estimating the hourly averaged U.S. background O₃ concentrations that flow into the risk models, the focus of our report is on better

understanding the limitations of the estimated concentrations resulting from the models that EPA used to estimate NA and or U.S. background O₃ levels.

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, 2012a), 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).

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. The authors noted the Yellowstone site 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 (US EPA, 2012a) 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 note that Lefohn et al. (2012-submitted) provided information that quantified the concentration enhancements associated with STE. Of specific relevance to the EPA and ISA (US EPA, 2012a), 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 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 O₃ concentrations in O₃ events exceeding or approaching air quality standards. Langford et al. (2011) found that surface O₃ measurements from 41 sampling stations indicate that ~13% of the variance in the maximum daily 8-h average (MDA8) O₃ 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 O₃ 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 O₃ over the entire area to ~55 ppb. This background was 10-15 ppb higher than the baseline O₃ in air transported ashore from the Pacific Ocean, and when combined with locally produced O₃, 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.

Work by Lin et al. (2012-submitted) has shown that the stratospheric contribution to the North American background (NAB) in the spring is larger than that from Asian transport (Fig. 1) based on high resolution run of the NOAA Geophysical Fluid Dynamics Laboratory AM3 model during the 2010 CalNex campaign. A unique feature of the GFDL global coupled atmosphere-oceans-land-sea ice model within a general circulation model is a fully coupled stratosphere-troposphere chemistry component. 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). 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, 2012a), 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 very 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 slightly modified results 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 (2012a) that annual coarse-resolution modeling applications on short-time scales have difficulty in characterizing observed O₃ concentrations.

3. Estimating North American Background Ozone Concentrations Using Empirical Data

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

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 1) and varies from year to year (i.e., 30 to 187 hours).

Table 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 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 has fallen in the range from 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 1) 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, 2012a) 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. It appears that O₃ concentrations associated with STT-S events supplement the enhanced O₃ concentrations that are associated with locally generated anthropogenic sources.

4. Conclusion

Correctly estimating hourly averaged US background O₃ concentrations is extremely important because characterizing NA background O₃ levels that are too low will result in (1) unreliable statistical significance outcomes associated with controlled laboratory exposure studies, (2) overestimated human health risk predictions, and (3) optimistic policy expectations of the levels to which hourly average O₃ concentrations can be lowered as a result of emission reduction requirements. The EPA points out in the ISA that currently all chemical transport models and not just the ones evaluated in the ISA have difficulty in predicting hourly and 8-h average concentrations on a day-specific basis. Recent work provides important evidence possibly explaining some of the reasons associated with the under predictions of NA background and U.S. background associated with the GEOS-Chem modeling. For example, strong evidence exists that STT processes are affecting surface O₃ concentrations at both high- and low-elevation monitoring sites across the US during the springtime, as well as other seasons. The enhancements associated with the STT process have an important effect on the estimation of hourly average U.S. background O₃ concentrations. The GEOS-Chem model predictions do not appear to be capturing the subtle, but important, enhancements in both the hourly average and monthly average 8-h average concentrations associated with STT-S processes. The modeling results reported by Lin et al. (2012-submitted) indicate that the background contribution is largest in the 50-80 ppb range. An implication from these results is that that the GEOS-Chem model underestimates both natural background and U.S. background at levels above 50 ppb.

Although the ISA (US EPA, 2012a) 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. Rather, 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). 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. The range of 3-year average of the 4th highest maximum 8-h average concentrations is from 50 to 52 ppb. When compared to the observational data at Trinidad Head, GEOS-Chem model predictions for the spring (US EPA, 2012a) 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.

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. Lefohn, Emery, and co-investigators indicate that hourly average total and background O₃ levels are under predicted. The results appear to confirm the EPA's conclusion in the ISA that the model used to estimate U.S. background may not be able to adequately capture the full impact of STE enhancements to surface O₃.

Results published in the literature indicate that the frequency of STE events that contribute to the variability of natural background is high. 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 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, 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 at Voyageurs NP (MN), Cuyahoga County (OH), and Chittenden County (VT) 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. 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 at Voyageurs NP, Cuyahoga County, and Chittenden County were 63, 77, and 83 ppb, respectively.

Besides STE 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.

As we indicate in our report, estimating background, as well as U.S. background O₃ concentrations correctly, is extremely important for assessing risk. 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. However, recent advancements in the science have begun to quantify the importance of stratospheric processes in affecting background and U.S. background surface O₃ concentrations. Based on our review of the science, we believe that these advancements can now allow the Agency to make improvements to its background O₃ estimates so that one of the important uncertainties associated with its risk assessments can be reduced.

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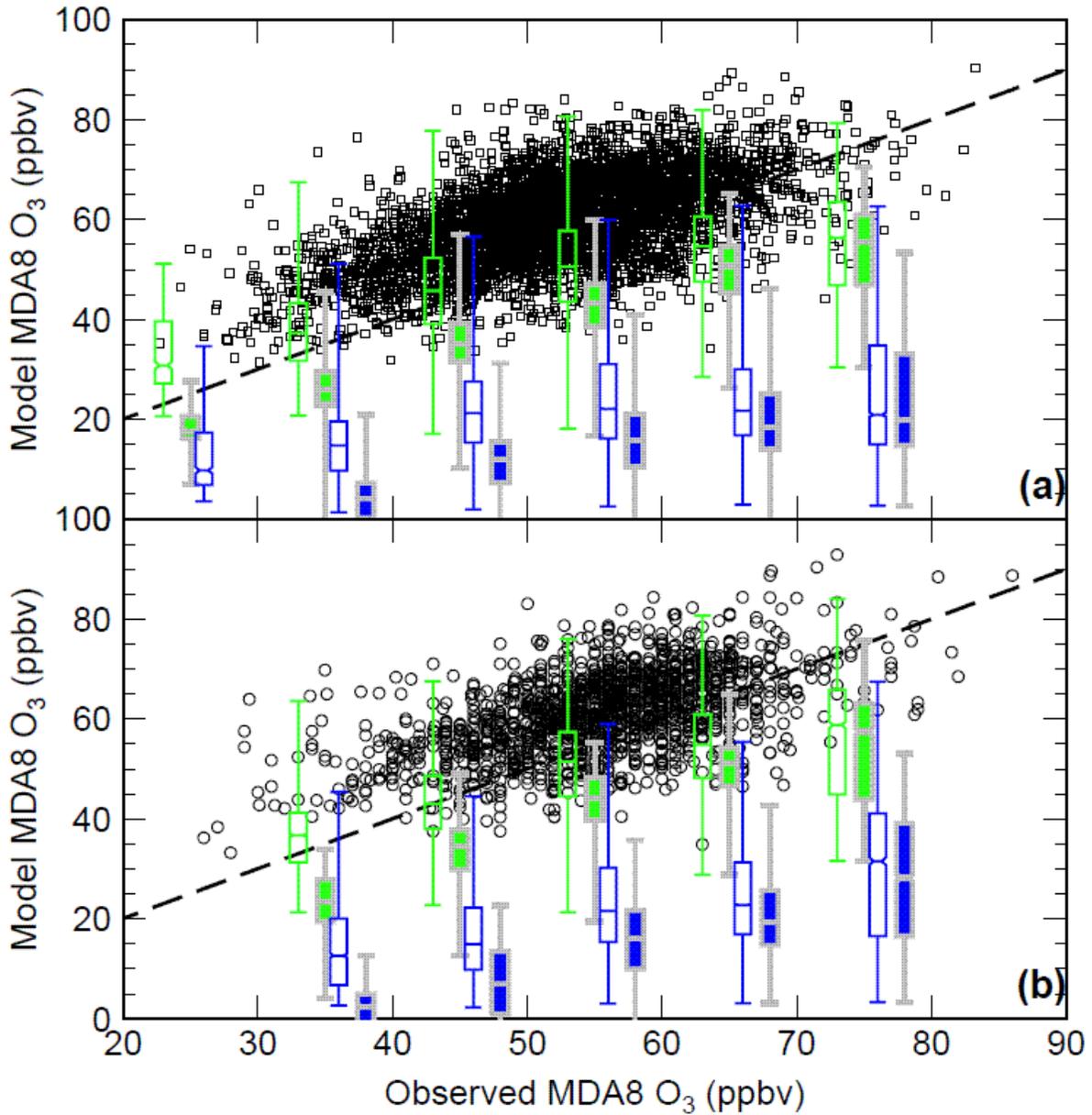


Fig. 1. Observed versus simulated MDA8 surface O₃ for April-June 2010 at (a) AQS sites over the central western U.S. and (b) 15 high-elevation sites. Also shown is the 1:1 line. The box-and-whisker plots (minimum, 25th, 50th, 75th percentiles, and maximum) give statistics of the NA background (green) 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).

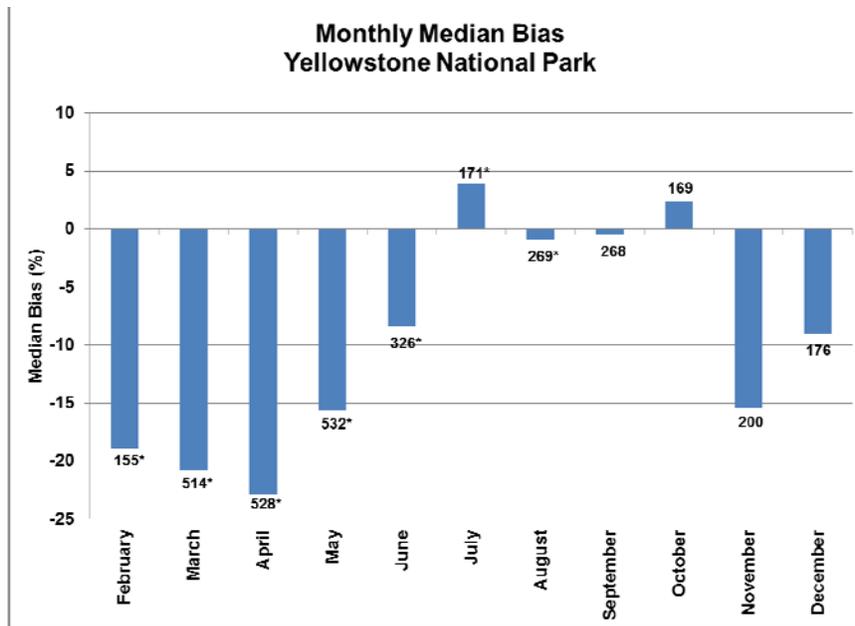


Fig. 2. Monthly median bias between modeled and observed hourly O₃ at Yellowstone NP for 2006. The total STT-S counts for each month are indicated; a statistically significant month is indicated with an asterisk (*).

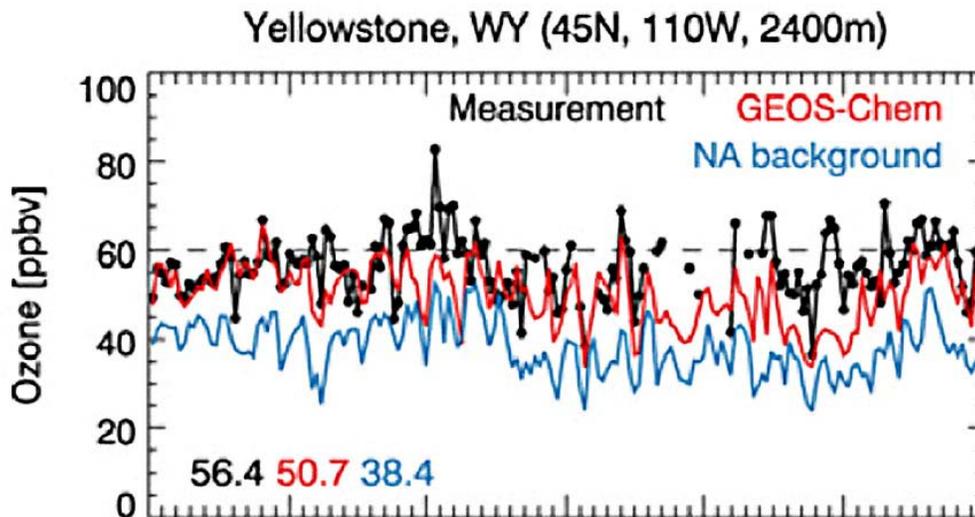


Fig. 3. Time series of measurements of daily maximum 8-h average ozone concentrations at Yellowstone NP with GEOS-Chem predictions for the base case and for the North American background case during March-August, 2006. Source: EPA (2012a).

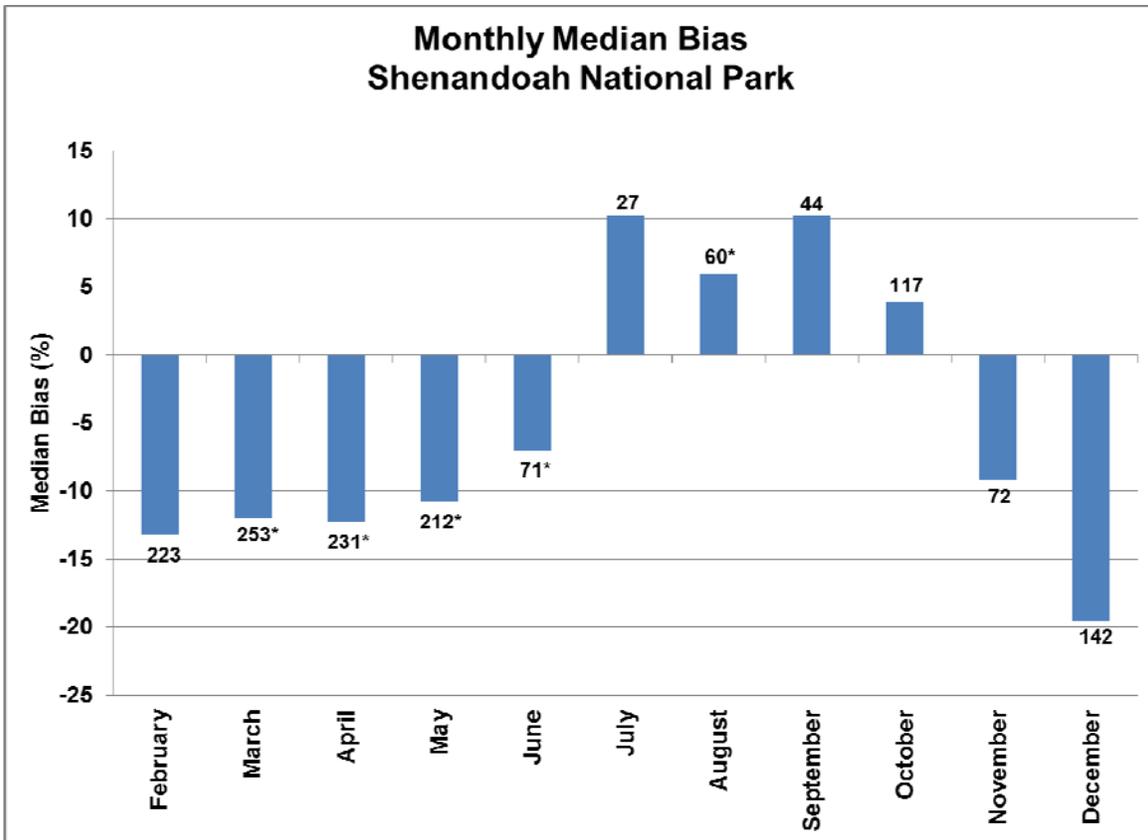


Fig. 4. Monthly median bias between modeled and observed hourly O₃ at Shenandoah National Park for 2006. The total STT-S counts for each month are indicated; a statistically significant month is indicated with an asterisk (*).



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.

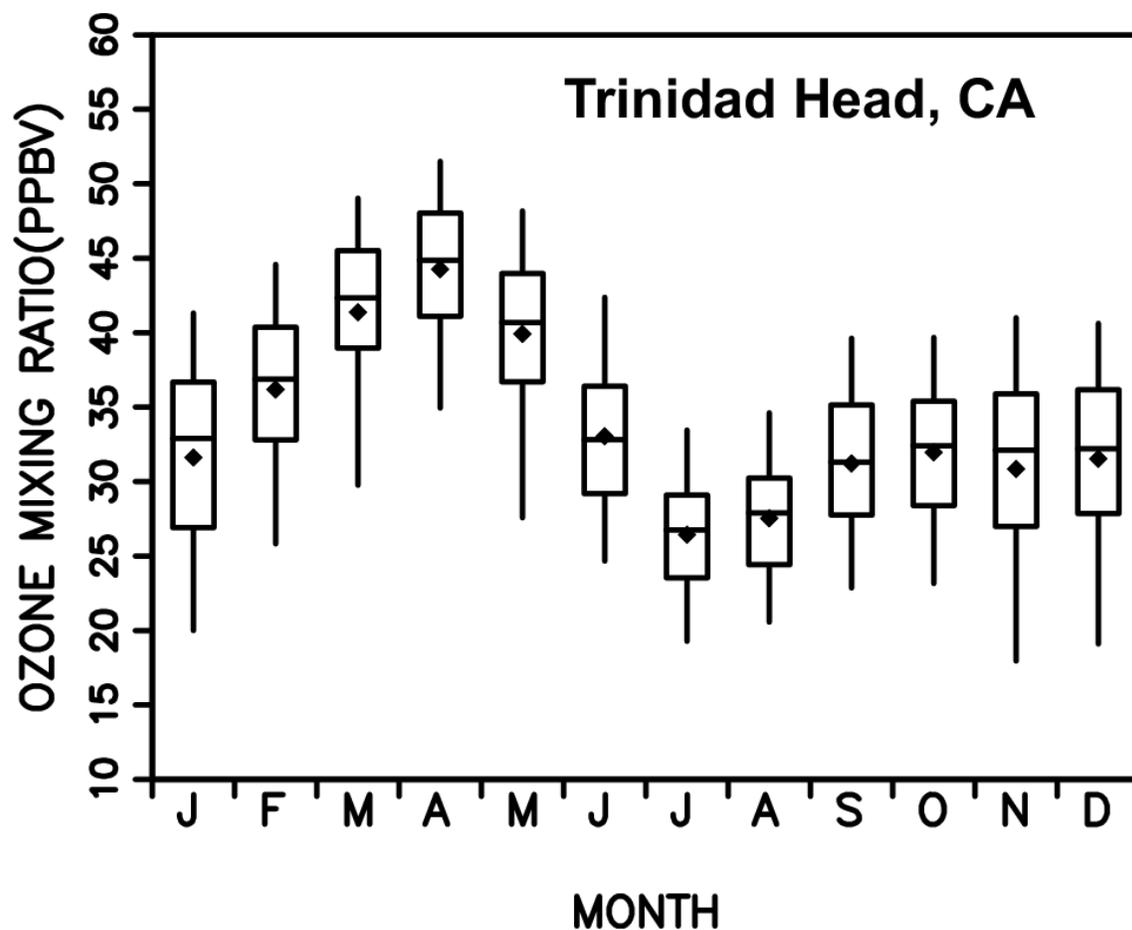


Fig. 6. Seasonal variation of daytime (1100-1800 Local Standard Time) surface O₃ at Trinidad Head, California (41N). The diamond is the mean, the horizontal bar within the box is the median, the box is the inner 50th percentile and the whiskers are the inner 90th percentile of the hourly averages based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

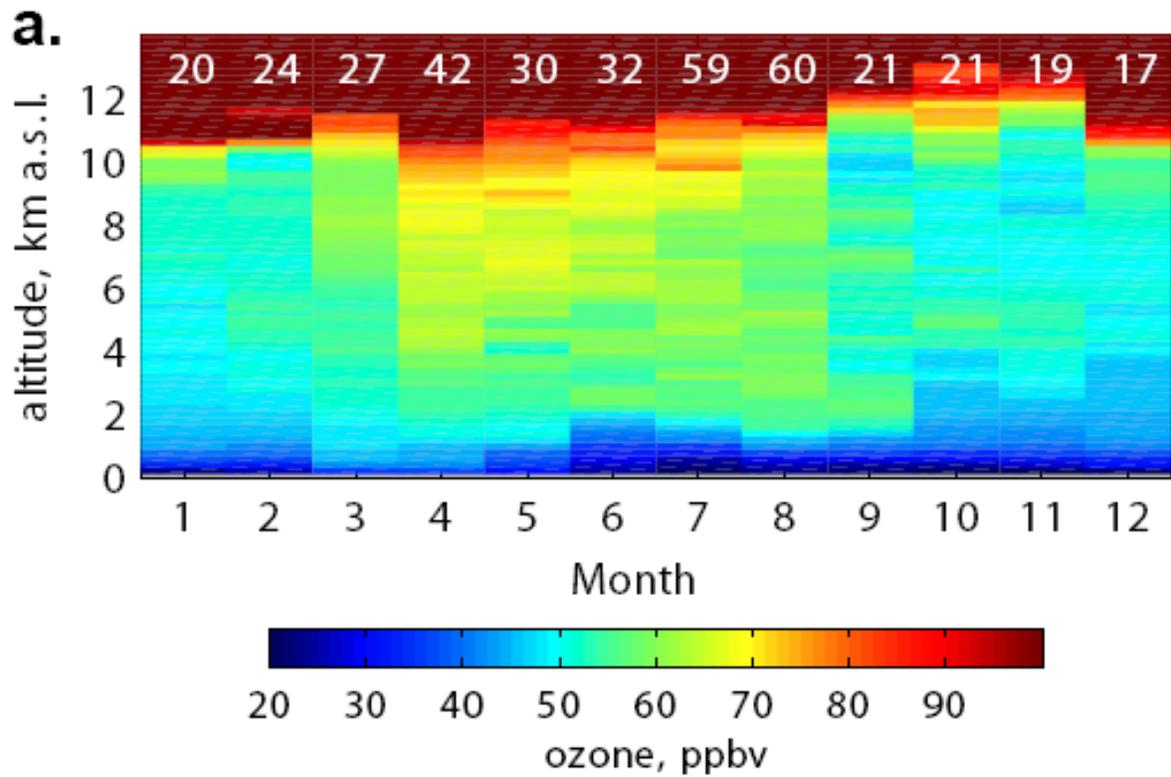


Fig.7. Tropospheric O₃ mixing ratios (ppbv) above the Trinidad Head Observatory from ozonsonde measurements. The numbers at the top of the figure for each month are the number of soundings for that month in the period 2004-2010. Source: Cooper et al. (2011).

Trinidad Head

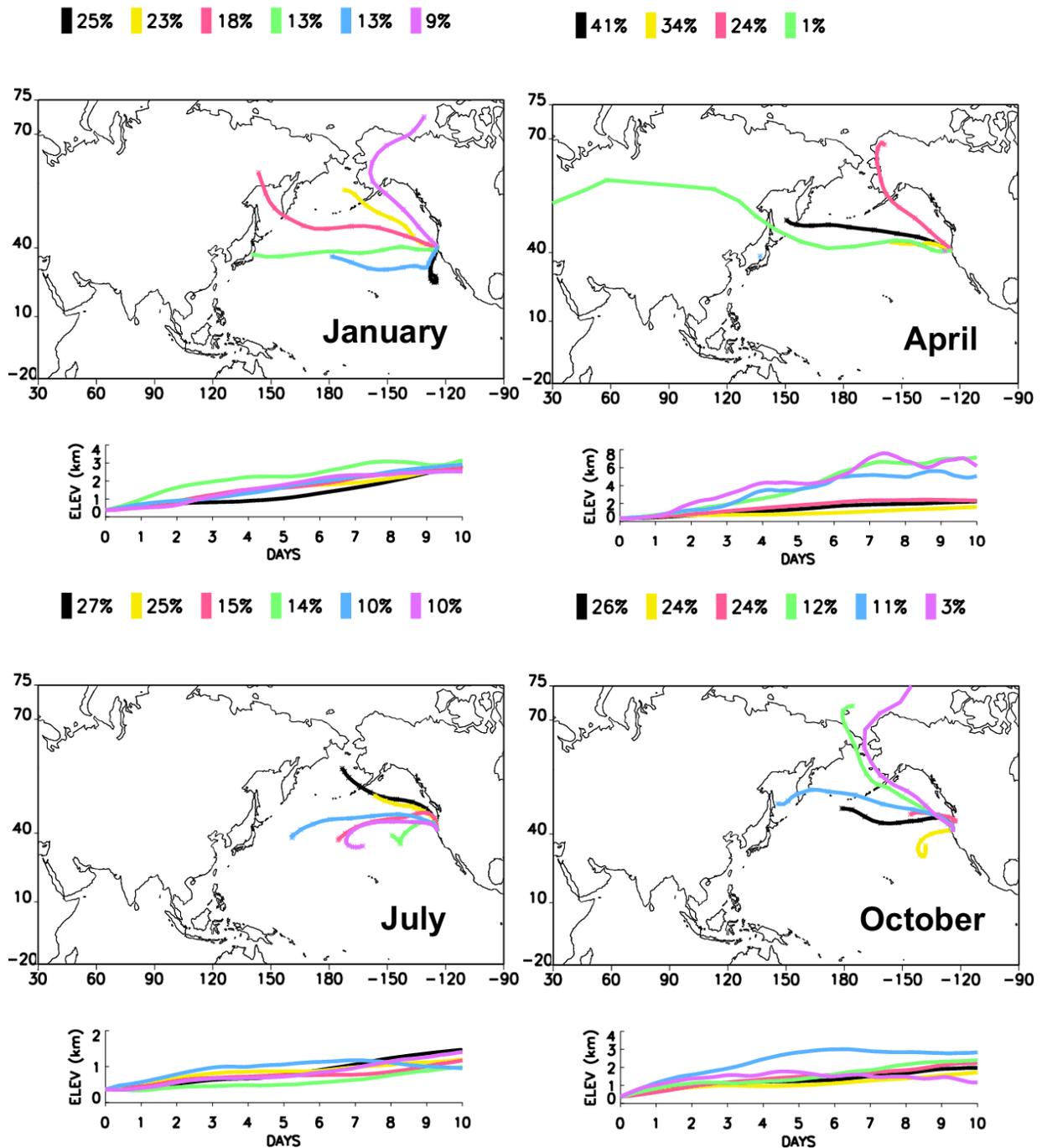


Fig. 8. Trajectory clusters for Trinidad Head, California for years 2002 to 2005 showing average flow characteristics at the station. Source: Oltmans et al. (2008).

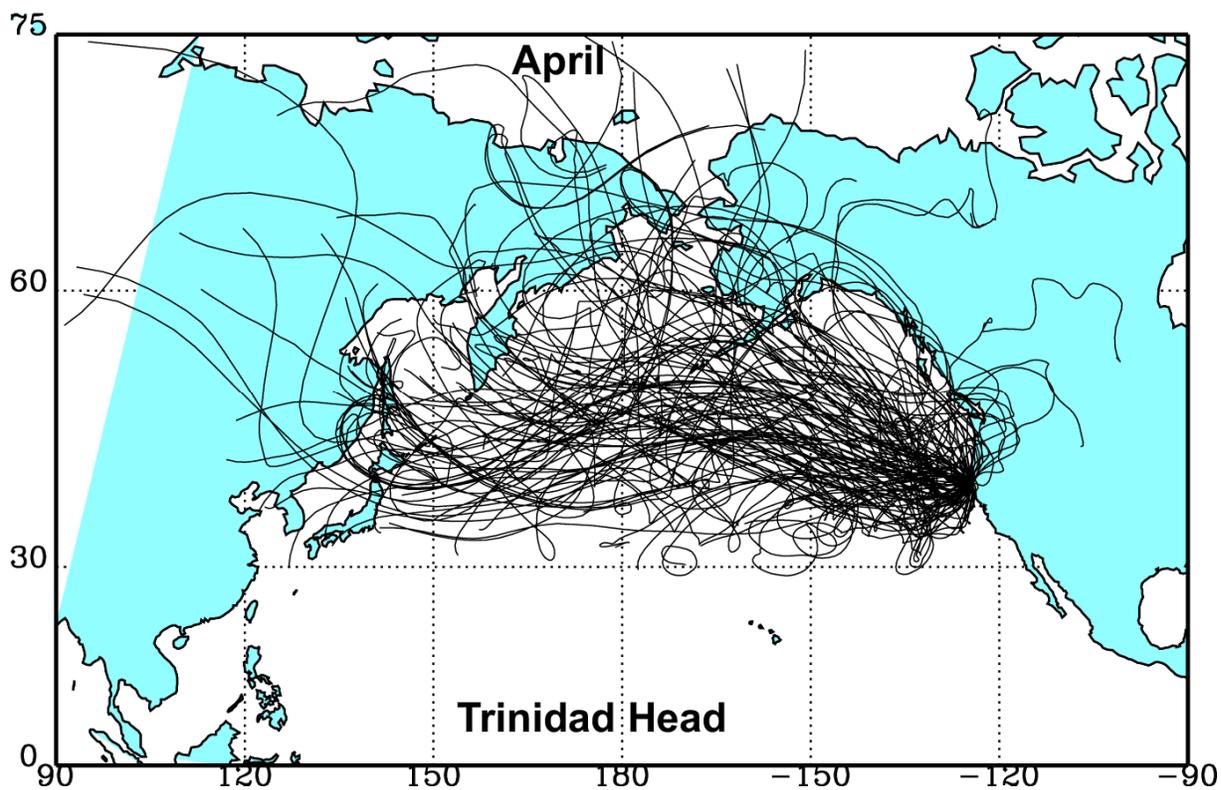


Fig. 9. Back trajectories from Trinidad Head, California on days with hourly average O_3 amounts ≥ 50 ppb for April in the years 2002 to 2005. Source: Oltmans et al. (2008).

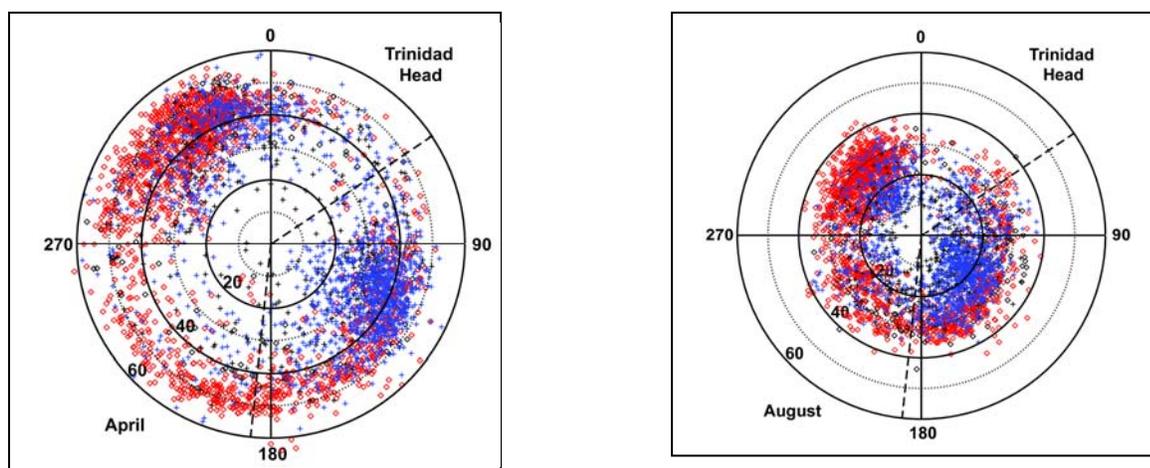


Fig. 10. Wind rose for O_3 mixing ratios at Trinidad Head for April and August for the period 2002-2007. The concentric circles mark the O_3 mixing from 0 ppbv at the center to 60 ppbv for the outer circle. The red diamonds are daytime (10-21 LST) values and the blue pluses are nighttime (22-09 LST) 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).

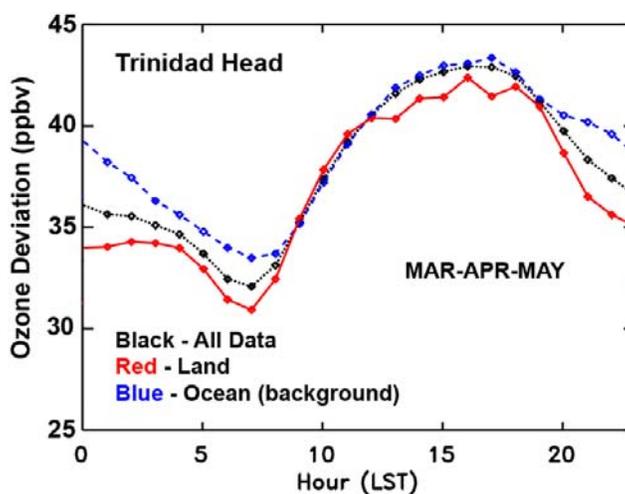


Fig. 11. Diurnal variation of O_3 mixing ratio at Trinidad Head for March-April-May for all observations (black), for observations when air reaching the observing site has recently come from over land (red), and for observations when air is coming from off the ocean (blue). Air reaching the site from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

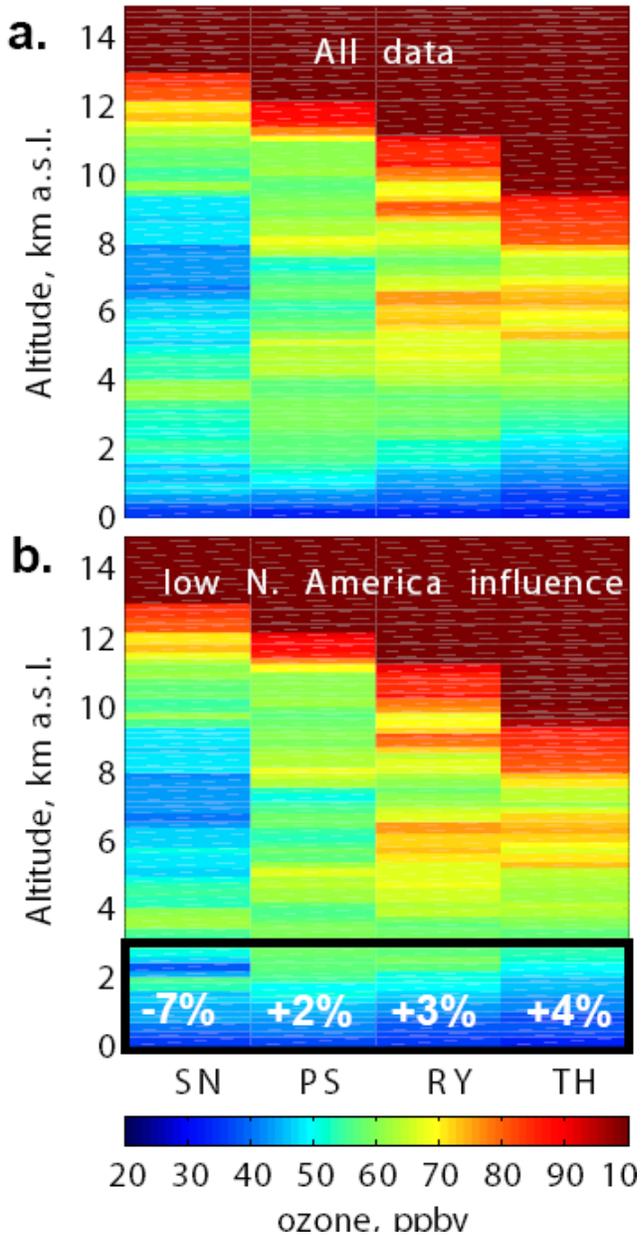


Fig. 12. a) Median O₃ profiles above the four coastal sites using all available data. b) Same as in a. but measurements with recent North American anthropogenic influence removed. Numbers in white indicate the percent change in the mass of O₃ in the 0-3 km range when air masses with strong North American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

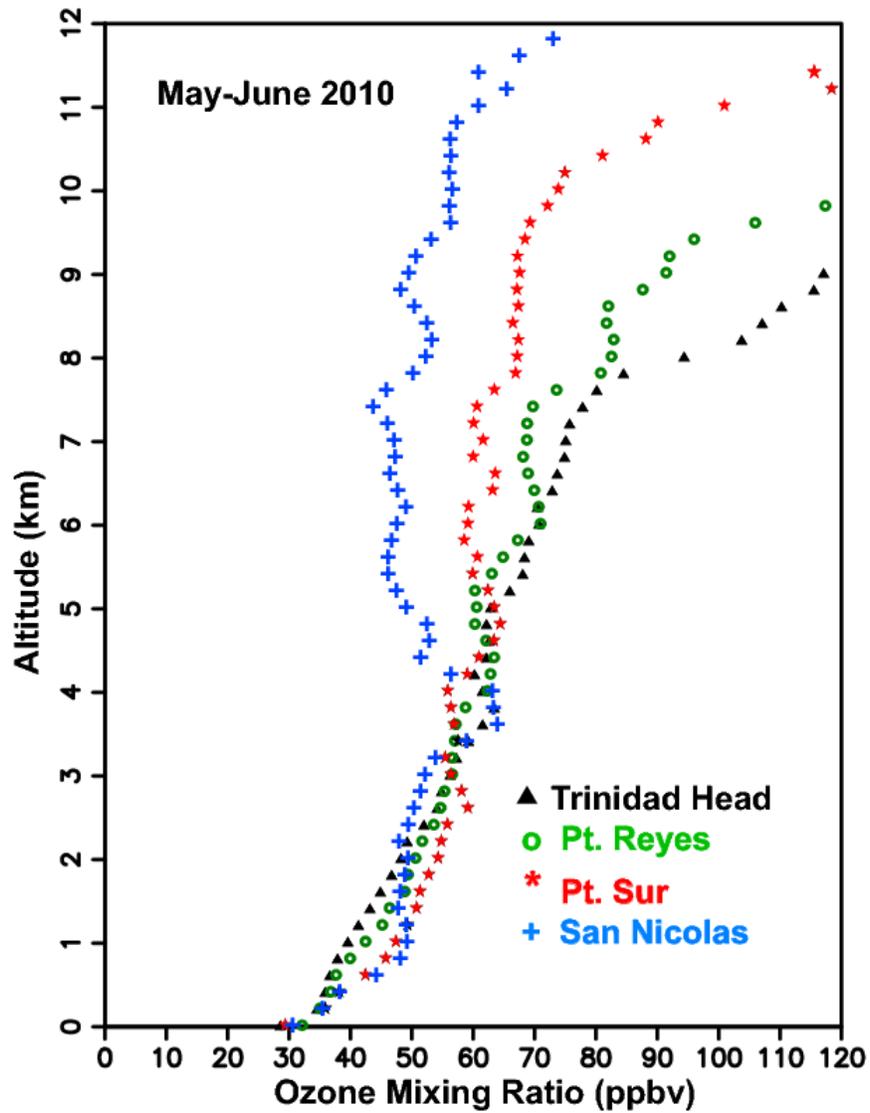


Fig. 13. Average O₃ mixing ratio profiles at four sites making ozonesonde observations during the IONS 2010/CalNex Campaign in May-June 2010. In the lower troposphere (<4 km) average O₃ amounts are similar along the California coast.

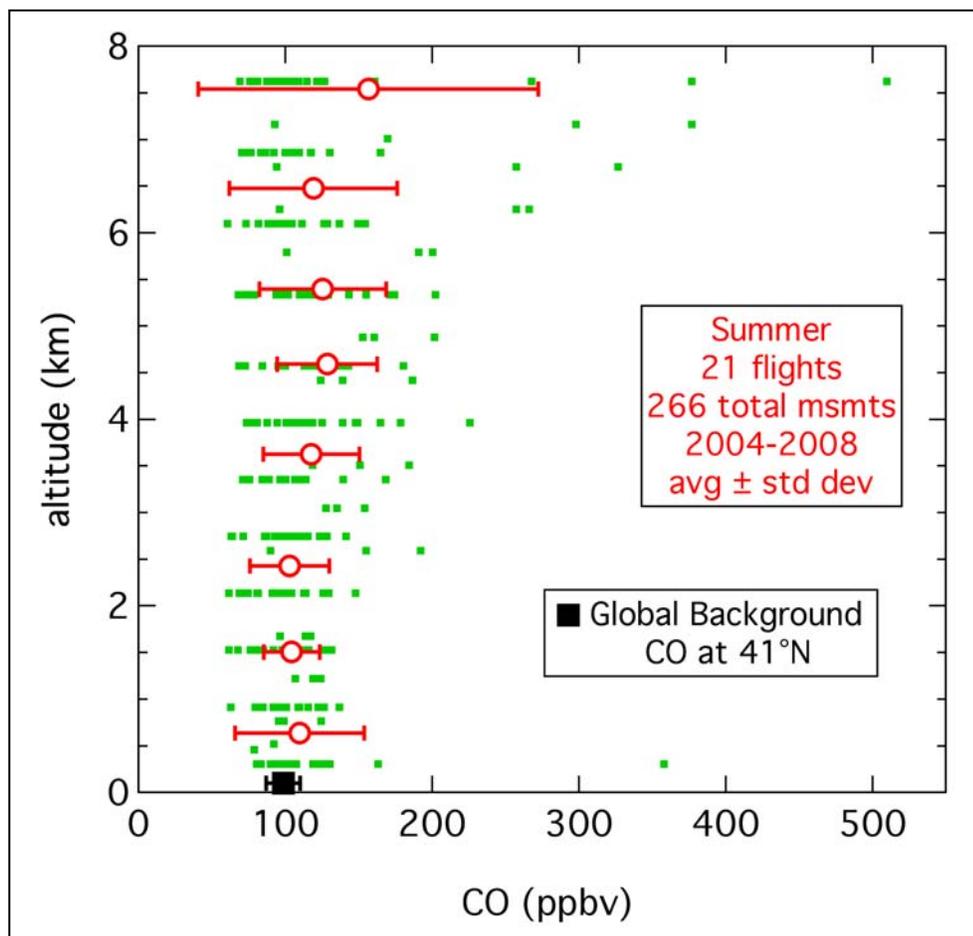
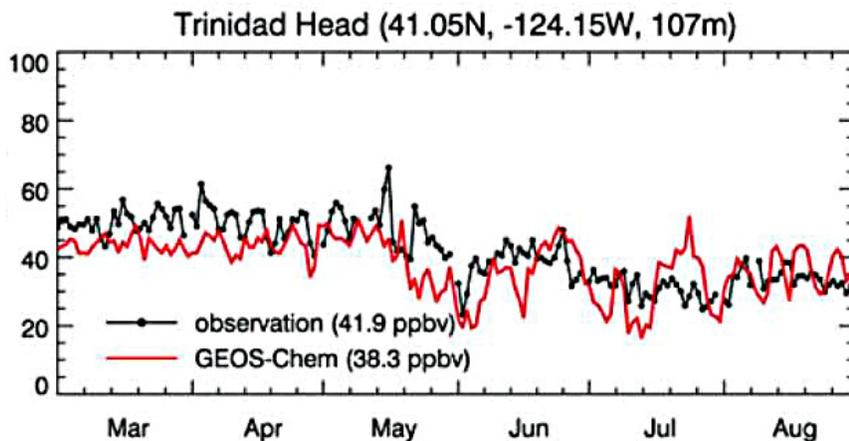


Fig. 14. Vertical profiles of carbon monoxide measured in flasks collected on aircraft flights above Trinidad Head CA. The green points give the individual measurements, and the red circles indicate averages and standard deviations for 1 km altitude segments. The black symbol gives the surface global 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)

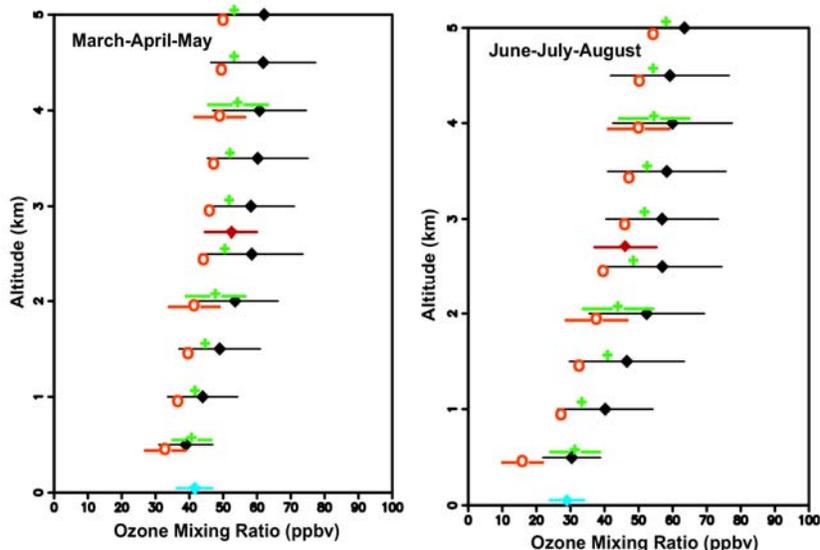


Fig. 15. a) Comparison of daily maximum 8-h average O_3 predicted using GEOS-Chem at $0.5^\circ \times 0.667^\circ$ with measurements at Trinidad Head, CA from March to August 2006. Source: US EPA (2012a). b). Average spring (March/April/May) O_3 profiles derived from ozone sondes at Trinidad Head are 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).

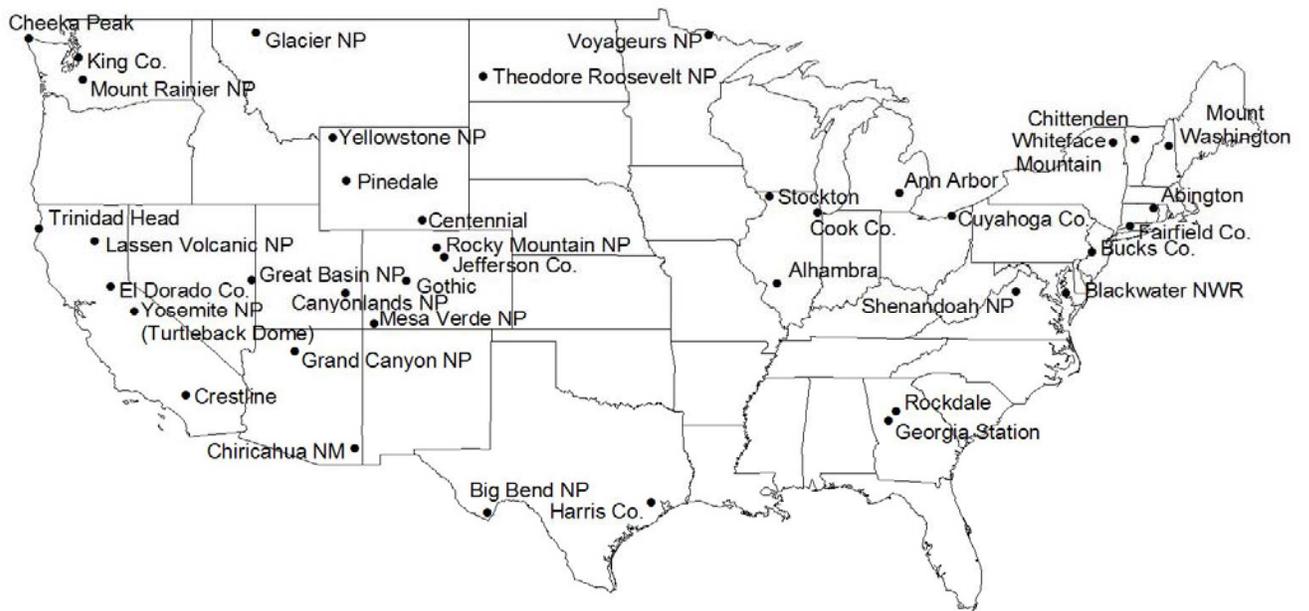


Fig. 16. Location of the 39 O₃ monitoring sites used in Lefohn et al. (2012-submitted) study.

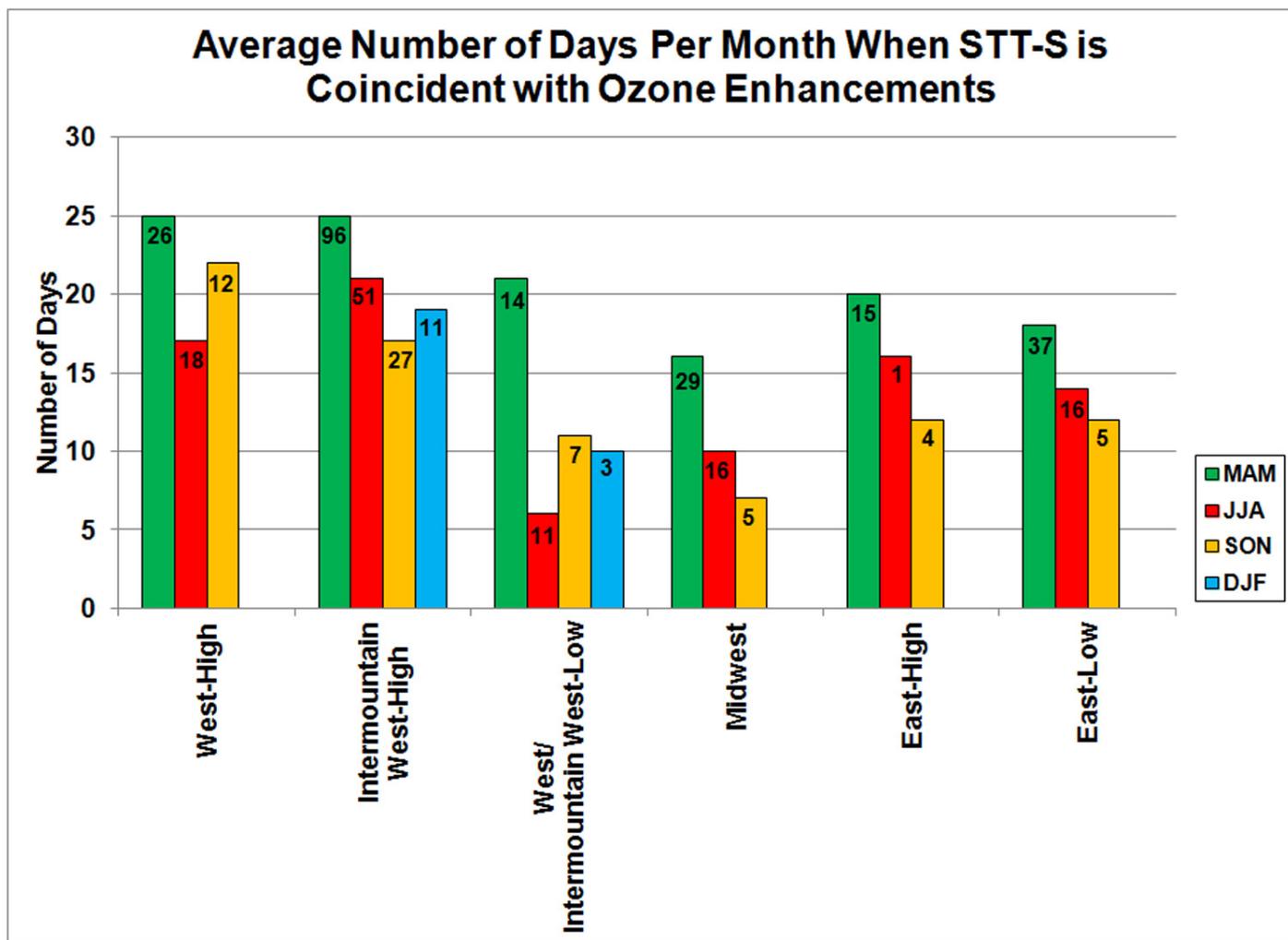


Fig. 17. Average number of days per month when STT-S is coincident with O₃ enhancements by geographic region for spring (March, April, and May), summer (June, July, and August), fall (September, October, and November), and winter (December, January, and February). The numbers within each bar are the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