Comments Relating to Section 3.4 (Policy-Relevant Background Concentrations) in the March 2011 Draft of the Integrated Science Assessment for Ozone and Related Photochemical Oxidants

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Executive Summary

EPA has defined Policy Relevant Background (PRB) as those concentrations that would result in the United States in the absence of anthropogenic emissions in continental North America (i.e., the United States, Canada and Mexico). PRB concentrations include contributions from natural sources everywhere in the world and from anthropogenic sources outside of North America. In 2007, the EPA used estimates of PRB O_3 concentrations in the exposure and risk assessments for 12 urban areas in the US. The PRB O_3 concentrations used in the exposure and risk assessments were based on monthly diurnal patterns of hourly average concentration predictions from the GEOS-Chem model for the 2001 April to September season. Correctly estimating hourly averaged PRB O_3 concentrations is very important because characterizing PRB O_3 levels that are too low can result in 1) overestimated human health risk predictions and 2) optimistic policy expectations of the levels to which hourly average O_3 concentrations can be lowered as a result of emission reduction requirements.

Meteorological evidence exists to support the observation that conditions representative of PRB are routinely encountered at the low-elevation monitoring site at Trinidad Head,

California. Trinidad Head regularly observes measurements under PRB 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 O_3 concentrations measured at this site. 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 PRB conditions. 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. The range of maximum hourly average concentrations for the month of April over the period 2002-2010 is 54 to 65 ppb. Generally, during April and May the monthly mean of the daily maximum 8-h average concentration is approximately 45 ppb, with the number of hourly average concentrations ≥ 50 ppb for April varying over the range of 30 to 187 for the period 2002 to 2010.

Eurasian emissions associated with biomass burning and their easterly transport contribute to O_3 concentrations at west coast O_3 monitoring sites under PRB conditions, as well as inland O_3 monitoring sites in Montana, Wyoming, and North Dakota. Published information indicates that biomass burning events in Eurasia are not infrequent. Naturally occurring stratospheric-tropospheric exchange (STE) processes contribute to background O_3 concentrations at the surface at both high and low-elevation monitoring sites. Natural processes, such as STE and biogenic emissions, contribute to the replenishment of PRB O_3 concentrations across the entire United States. Indications at many of the lower-elevation monitoring sites investigated using a Lagrangian trajectory model (LAGRANTO) are that numerous days occur in which a strong relationship exists between enhanced O_3 concentrations and stratosphere-to-troposphere transport that reaches the surface (STT-S) particularly during the springtime. When statistically

significant coincidences occurred at the lower elevation sites, the daily maximum hourly average concentrations were mostly in the 50-65 ppb range; the daily maximum 8-h average concentrations were usually in the 50-62 ppb range.

Evidence exists that background O_3 varies strongly from year-to-year and that the background contribution at the surface in the western US is larger than current models indicate. The GEOS-Chem model appears to underestimate the frequently occurring hourly average PRB O_3 concentrations $\geq$ 50 ppb experienced during the springtime at the west coast of the US. In addition, the model does not adequately capture the replenishment processes associated with STT events that reach the surface at both high- and low-elevation monitoring sites and the wildfires that enhance PRB O_3 hourly average concentrations.

The GEOS-Chem model estimates that the mean value for PRB O_3 for April-May in the vicinity of the low-elevation site at Trinidad Head for 2001 is below 35 ppb. Generally at Trinidad Head, the mean value is approximately 45 ppb for April and May, with the number of hourly average concentrations $\geq$ 50 ppb for April varying over the range of 30 to 187 for the period 2002 to 2010. For the Vermont Area, GEOS-Chem appears to predict for the April-May period a mean value for PRB O_3 below 35 ppb (page 3-34). For the low-elevation Chittenden County Vermont site, we report a statistically significant relationship between STT-S and enhanced hourly average concentrations $\geq$ 50 ppb for April 2006 (19 days), April 2008 (20 days), and May 2008 (17 days). The monthly mean of the daily maximum 8-h average concentrations for these periods was 49 ppb, 54 ppb, and 46 ppb, respectively suggesting that in April-May 2008, for example, the contribution from the natural background may be elevated above a model-determined value for a single year.

Time series plots of 8-h daily max O_3 concentrations observed at 14 CASTNET sites are illustrated in the Integrated Scientific Assessment. The time series for the various sites (e.g., Acadia NP,

Crockett, Coffeeville, Voyageurs NP, Rocky Mountain NP, Yellowstone NP, Centennial, Canyonland NP, Mount Rainier NP, Lassen Volcanic NP, and Pinnacles NM) appear to show a pattern of lower estimation of 8-h daily maximum concentrations than actual values at times during the spring when O_3 is measured under conditions that frequently are representative of PRB. The lack of agreement may be associated with the model's inability to capture the O_3 concentration enhancements under these elevated PRB O_3 level conditions.

Although the EPA has attempted to summarize the science of PRB O_3, it is our opinion that further discussion is required in the document to adequately assess the importance of the various PRB processes and their affects on defining PRB O_3 hourly average concentrations. Important contributions, although cited in previous versions of the Criteria Document, such as the important contribution by Ludwig et al. (1977), are omitted in the current version. Evidence exists that frequently occurring STT processes are affecting springtime surface O_3 concentrations at both high- and low-elevation monitoring sites and the enhancements associated with the STT process have an important effect on estimating hourly average PRB O_3 concentrations.

1. Introduction

When the EPA proposes ranges for the primary O_3 standard it bases its estimates on a combination of (1) human laboratory experimental results and (2) risk assessments that incorporate the assumptions of the modeled estimates for policy-relevant background (PRB) O_3 daily afternoon concentrations. EPA has defined PRB as those concentrations that would result in the United States in the absence of anthropogenic emissions in continental North America (i.e., the United States, Canada and Mexico) (U.S. EPA, 2011, page 3-25 – 3-26). Policy Relevant Background concentrations include contributions from natural sources everywhere in the world and from anthropogenic sources outside of North America. Contributions to PRB O_3 include: (a) photochemical interactions involving natural emissions of VOCs, NO_x, and CO; (b) the long-range transport of O_3 and its precursors from outside North America; and (c) stratospheric-tropospheric exchange (STE). Natural sources of O_3 precursors include biogenic emissions, wildfires, and lightning. Biogenic emissions from agricultural activities are not considered in the formation of PRB O_3.

For the EPA review of the O_3 human health standards completed in 2008 (US EPA, 2006; 2007), the EPA determined that PRB O_3 concentrations could not be derived solely from measurements of O_3 at relatively remote monitoring (RRMS) sites because of long-range transport from anthropogenic source regions within North America. EPA estimated PRB O_3 concentrations using the global chemical transport model, GEOS-Chem. The EPA (2006; 2007) determined that 4-hour average PRB O_3 concentrations at the surface were generally in the range of 15 to 35 ppb in the afternoon, and declined under conditions conducive to high O_3 episodes. PRB O_3 concentrations were estimated to be highest during spring, due to contributions from hemispheric pollution and stratospheric intrusions, and were predicted to decline into summer.

The stratospheric contribution to surface O_3 was estimated to typically be below 20 ppb and was more frequently elevated at high-altitude than low-altitude sites (US EPA 2006; 2007).

Estimates of PRB O_3 concentrations were used in the exposure and risk assessments for 12 urban areas (Atlanta, Boston, Chicago, Cleveland, DC, Detroit, Houston, Los Angeles, New York, Philadelphia, Sacramento, and St. Louis) in the US. The PRB estimates were based on monthly diurnal patterns of hourly average concentration predictions of the model described in Fiore et al. (2003) for the 2001 April to September season (US EPA, 2007).

As outlined by the US EPA (2007) in its Staff Paper, the basic structure of the risk assessments reflects the two different types of human studies on which the O_3 health risk assessment is based:

- Controlled human exposure studies; and
- Epidemiological studies.

Controlled human exposure studies involve volunteer subjects who are exposed while engaged in different exercise regimens to specified levels of O_3 under controlled conditions for specified amounts of time. For its 2007 health risk assessment, the EPA used probabilistic exposure-response relationships based on analysis of individual data that described the relationship between a measure of personal exposure to O_3 and measures of lung function recorded in the studies. The measure of personal exposure to ambient O_3 was typically some function of hourly exposures (i.e., 1-hr maximum or 8-hr maximum concentrations). Therefore, a risk assessment based on exposure-response relationships derived from controlled human exposure study data required estimates of personal exposure to ambient O_3, typically on a 1-hr or multi-hour basis. Because data on personal hourly exposures to O_3 of ambient origin were not available, estimates of personal exposures to varying ambient concentrations were derived

through exposure modeling. In contrast to the exposure-response relationships derived from controlled human exposure studies, epidemiological studies provided estimated concentration-response relationships based on data collected in real world community settings. Ambient O_3 concentrations, measured as the average of monitor-specific measurements, using population oriented monitors, were used as a surrogate measure of population exposure.

The US EPA (2007-page 5-13) noted that the risk estimates developed for both the recent air quality scenario and just meeting the current 8-h standard represent risks associated with O_3 levels in excess of estimated PRB O_3 concentrations. The results of the global tropospheric O_3 model GEOS-Chem were used to estimate average background O_3 levels for different geographic regions across the US. These GEOS-Chem simulations included a background simulation in which North American anthropogenic emissions of NO_x, non-methane volatile organic compounds, and carbon monoxide were set to zero, as described in Fiore et al. (2003). As indicated above and in US EPA (2007, page 5-22), the Agency estimated monthly background diurnal concentrations for each of 12 urban areas based on the GEOS-Chem simulations (US EPA, 2007 - Appendix 2-A).

Health risk assessment based on controlled human exposure data

For the health risk assessment based on data from controlled human exposure studies exposure estimates for a number of different air quality scenarios were combined with probabilistic exposure-response relationships derived from the controlled human exposure studies to develop risk estimates associated with recent air quality and just meeting the current and alternative standards in excess of PRB. Hourly average concentrations were used as inputs into the risk models.

A population risk estimate for a given lung function decrement (e.g., ≥15% change in FEV_1) was an estimate of the expected number of people who would experience that lung function decrement. Since EPA was interested in risk estimates associated with hourly O_3 concentrations in excess of PRB concentrations, the following steps were taken to estimate the controlled human exposure risk associated with recent conditions in excess of PRB:

- Expected risk given the personal exposures associated with recent ambient O_3 concentrations was estimated.
- Expected risk given the personal exposures associated with estimated PRB ambient O_3 concentrations was estimated.
- The latter was subtracted from the former.

The population risk was then calculated by multiplying the resulting expected risk by the number of people in the relevant population.

Health risk assessment based on epidemiological data

For estimating the risks associated with epidemiological data, the estimate of the change in incidence of a health effect associated with a change in O_3 concentrations from recent levels to PRB levels in an assessment location was required. Two time series of O_3 concentrations were required for each location: (1) hourly O_3 concentrations from a recent year for the period April 1 through September 30, and (2) hourly PRB O_3 concentrations for the same time period. In order to be consistent with the approach generally used in the epidemiological studies that estimated O_3 concentration-response functions, the (spatial) average ambient O_3 concentration on each hour for which measured data were available is deemed most appropriate for the epidemiological risk assessment. A composite monitor data set was created for each assessment location based on averaging each hourly value from all monitors eligible for comparison with the current standard

for each hour of the day. Table 4-7 provides a summary of the design values for the 12 urban study areas.

Different exposure metrics have been used in epidemiological O_3 studies, including the 24-hr average and the daily 1-hr and 8-hr maximum. Therefore, daily changes at the composite monitor in the O_3 exposure metric appropriate to a given concentration-response function were calculated for use in the risk assessment. For example, if a concentration-response function related daily mortality to daily 1-hr maximum O_3 concentrations, the daily changes in 1-hr maximum O_3 concentrations at the composite monitor were calculated. In the first part of the epidemiology-based risk assessment, in which risks associated with the recent levels of O_3 above PRB levels were estimated, this required the following steps:

- Using the monitor-specific input streams of hourly O_3 concentrations from a recent year, calculate a stream of hourly O_3 concentrations at the composite monitor. The recent O_3 concentration at the composite monitor for a given hour on a given day is the average of the monitor-specific O_3 concentrations for that hour on that day.

- Using this stream of hourly O_3concentrations from a recent year at the composite monitor, calculate the 1-hr maximum O_3 concentration for each day at the composite monitor.

- Using the monitor-specific input streams of hourly background O_3 concentrations, calculate a stream of hourly PRB O_3 concentrations at the composite monitor.

- Using this stream of PRB hourly O_3 concentrations at the composite monitor, calculate the 1-hr maximum PRB O_3 concentration for each day at the composite monitor.

- For each day, calculate Δx = (the 1-hr maximum O_3 concentration for that day at the composite monitor) - (the 1-hr maximum PRB O_3 concentration for that day at the composite monitor).

Similarly, calculations for concentration-response functions that used a different exposure metric (e.g., the 8-hr daily maximum or 24-hr average) were done analogously, using the exposure metric appropriate to the concentration-response function.

Clearly, correctly estimating hourly averaged PRB O_3 concentrations is very important because characterizing PRB O_3 levels that are too low can result in 1) overestimated human health risk predictions and 2) optimistic policy expectations of the levels to which hourly average O_3 concentrations can be lowered as a result of emission reduction requirements.

2. Estimating policy-relevant background ozone concentrations using empirical data

2.1 The influence of PRB O_3 concentrations at Trinidad Head (CA)

Although PRB O_3 is not a directly measureable quantity, conditions representative of PRB are routinely encountered at the monitoring site at Trinidad Head, CA. Trinidad Head 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. 1). The site is connected to the mainland only on its northern end. Surface O_3 measurements began in mid-April 2002 and continue to the present at Trinidad Head. In addition O_3 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 O_3 throughout the troposphere at Trinidad with a maximum in April and May (Figs. 2 and 3). Long-range transport outside of North America and natural processes such as stratospheric enhancement contribute to O_3 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 PRB conditions. 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 (i.e., 30 to 187 hours). The range of maximum hourly average concentrations for the month of April over the period 2002-2010 is 54 – 65 ppb. For the period 2002-2010, Table 2 summarizes the hourly percentiles, number of hourly occurrences ≥ 50 ppb, SUM06 (the sum of hourly average concentrations ≥ 60 ppb), and the W126 cumulative sigmoidally weighted (see Lefohn and Runeckles, 1987; Lefohn et al., 1988) exposure indices that were experienced at Trinidad Head. The units of the SUM06 and W126 exposure indices are ppm-h. In the period 2003-2010, daily maximum 8-h average O_3 concentrations exceeded 60 ppb during the springtime. Figs. 4 and 5 illustrate the range of daily maximum 8-h average concentrations for 2003 and 2006, respectively and compare this range with the EPA-defined policy-relevant background O_3 concentrations of 15 to 35 ppb. Trinidad Head in 2003 experienced greater numbers of hourly average O_3 concentrations ≥ 50 ppb in comparison to 2006. A 3-year average of the 4th highest maximum 8-h average concentrations falls in the range from 50 to 52 ppb. These relatively high values at a marine boundary layer location under conditions representative of PRB occur during the springtime.

Trinidad Head regularly observes measurements under PRB conditions for daytime observations (i.e., mid morning to late afternoon). This has been demonstrated by a number of studies (Oltmans et al., 2008; Goldstein et al., 2004; Parrish et al., 2010; Huang et al., 2010; Cooper et al., 2011). Oltmans et al. (2008) used both a trajectory climatology (cluster analysis) (Fig. 6) as well as individual trajectories for O_3 events (Fig. 7), where O_3 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 PRB conditions. In addition, Oltmans et al. (2008) analyzed the local wind conditions for each O_3 hourly average (Fig. 8) and showed that high O_3

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 O_3 deposition and possible chemical loss. The study of Goldstein et al. (2004), which made detailed chemical measurements during a campaign in April and May 2002, used 3 ppt MTBE (a trace additive in gasoline) as a threshold to separate between minimal local influence (60% of the observations) and recently polluted air (40% of observations) to filter out significant influences from North American continental emissions. This included both day and night observations implying that a large fraction of measurements with continental influence were nighttime observations coming with the offshore (i.e., from the land) winds. Under these circumstances, the O_3 concentrations would not be representative of PRB conditions and have significantly lower O_3 amounts (see Fig. 8) as noted by Goldstein et al. (2004), Parrish et al. (2009), and Cooper et al. (2011). 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 O_3 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 9). 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 California coast, have very low exposure to local emissions and mid-afternoon (the time of the soundings) exposure to marine air masses.

It is not clear that O_3 observations under conditions that are informative of PRB O_3 levels should have a diurnal variation. At Trinidad Head the diurnal variation reflects the influence of the heating and cooling of the nearby continental land mass that induces a local land/sea breeze circulation when the prevailing synoptic flow does dominate. At relatively remote island

locations that we have studied, such as Bermuda (Fig.10), the prevailing flow pattern is not normally interrupted by a local wind circulation and there is essentially no difference between day and night O_3 amounts. This suggests that air entering the coast of North America under conditions representative of PRB likely has O_3 levels that are best represented by the daytime values at a location such as Trinidad Head.

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 observation, Cooper et al. (2011) concluded that the median O_3 profiles above these sites are representative of baseline O_3 along the U.S. west coast (Fig. 11). In the lower troposphere, the median O_3 values at these sites are very similar indicating that the O_3 distribution is dominated by the broad scale flow off the Pacific Ocean. The strong similarity in lower troposphere O_3 among all of the sites suggests that the O_3 values at Trinidad Head are indicative of O_3 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. 12). 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 O_3 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 O_3 as well as CO.

Detailed analysis of surface and profile O_3 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 Policy Relevant Background O_3. In the spring at a marine boundary layer site, such as Trinidad Head, hourly average O_3 concentrations are often ≥ 50 ppb and in April, the monthly average daytime maximum is ~45 ppb. Although marine boundary layer O_3 decreases through the summer, air flowing into California above the boundary layer remains on average above 50 ppb through the summer. There is little evidence that recent North American emissions contribute significantly to O_3 measured at coastal sites. In fact, as indicated above, such emissions result in depressed O_3 concentrations levels.

2.2 Long-range transport effects on PRB in the western US from Eurasian biomass burning

Eurasian emissions associated with biomass burning and their easterly transport contribute to O_3 concentrations at west coast O_3 monitoring sites under PRB conditions, as well as inland O_3 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 O_3 enhancements in surface O_3 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 O_3 readings were recorded in April 2008 at surface O_3 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 O_3 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 O_3 readings exceeding 50 ppb were experienced during this period. As the biomass burning enhanced O_3 plume moved further into the interior of the US between 18-20 April through a northern tier of states (Montana, Wyoming, North Dakota), surface O_3 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 O_3 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 O_3 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 O_3 amounts at Trinidad Head in April 2003 as well (Oltmans et al., 2010).

Fig. 13 illustrates the 3-year average of the fourth highest annual daily maximum 8-h average concentrations for four background O_3 monitoring sites for the 2008-2010 period that

appear to have been influenced in April 2008 by a PRB component that was associated with the long-range transport of emissions from Eurasian biomass burning.

2.3 The importance of the stratosphere in influencing surface ozone concentrations

Natural processes, such as stratospheric-tropospheric exchange (STE) and biogenic emissions, contribute to the replenishment of PRB O_3 concentrations across the entire United States (U.S. EPA, 2006). Naturally occurring STE processes contribute to background O_3 concentrations at the surface at both high and low-elevation monitoring sites (e.g., Ludwig et al., 1977; Haagenson et al., 1981; Schuepbach et al., 1999; Stohl et al., 2000; Lefohn et al., 2001; Cooper et al., 2005; Hocking et al., 2007; Ordóńez et al., 2007; Langford et al., 2009). Hourly average concentration ≥ 0.05 ppm in the springtime can be attributed at times to these natural processes (Lefohn et al., 2001; Cooper et al., 2005; Langford et al., 2009). Key to the importance of STE processes in replenishing PRB is the frequency of STE events. Indications of the importance of the frequency of STE events affecting lower tropospheric O_3 concentrations have been reported (Ordóńez et al., 2007). Reed (1955) and Danielsen (1968) used instrumented high-altitude aircraft measurements of radioactivity and O_3 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 O_3 by characterizing the behavior of radioactive debris injected into the stratosphere during nuclear weapons testing in the 1960s. Using ^{90}Sr, Ludwig et al. (1977) concluded that a significant stratospheric contribution to ground level O_3 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.

At some monitoring sites, the stratospheric contribution to surface O_3 can be significant and can lead to exceedances of the 0.075 ppm 2008 NAAQS O_3 standard. During 1999, STE contributed to one of the 4 highest daily 8-h maxima at one or more surface sites in the metropolitan Denver area on at least 4 occasions during the March–October O_3 season (Langford et al., 2009).

To explore the frequency of stratosphere-to-troposphere transport (STT) events enhancing hourly average O_3 concentrations, our international research team's most current results shows the importance of STT events at both high- and low-elevation monitoring sites. The Lagrangian Analysis Tool (LAGRANTO) trajectory model (Wernli and Davies, 1997) was used to identify specific days when stratosphere-to-troposphere transport was optimal to enhance surface O_3 levels. The STT events were identified with the method introduced by Wernli and Bourqui (2002) and later applied by Sprenger and Wernli (2003) and Stohl et al. (2003) for compiling multi-year climatologies of such events. Lefohn et al. (2011) explored the statistically significant coincidences between the number of days with a daily maximum hourly average O_3 concentration $\geq$ 50 ppb and stratosphere-to-troposphere transport to surface (STT-S). In evaluating 12 O_3 monitoring sites across the US, we found that the high-elevation site at Yellowstone National Park in Wyoming (2468 m asl) exhibited the most statistically significant coincidences (i.e., more than 19 days a month) during the spring and summer for hourly average O_3 concentrations $\geq$ 50 ppb for the 2006 to 2008 period. Fig. 14 illustrates the frequency of the stratosphere-to-troposphere transport events to the surface at Yellowstone NP for the period. The most frequent occurrence of STT –S events were during the springtime months. At this site, the maximum hourly springtime average O_3 concentrations were usually in the 60-70 ppb range. The maximum daily 8-h average concentrations mostly ranged from 50 to 65 ppb. However, there

were instances when the maximum hourly average concentrations associated with STT-S events were greater than 80 ppb (e.g., May 2, 2006). Our detailed case-study analysis indicated that this specific O_3 concentration enhancement can be regarded as a classical STT event associated with tropopause folding and the formation of stratospheric PV streamers. The range of the 3-year average of the 4th highest 8-h daily maximum values at Yellowstone National Park is from 63 to 67 ppb. For the period 1997-2008, 75% of the top four daily maximum 8-h average concentrations occurred during the springtime. The EPA has indicated that Yellowstone National Park would violate the primary O_3 standard for the 3-year 2006-2008 period if the level were set at 0.065 ppm. Based on our research results and the results published by others, natural processes, such as stratospheric-tropospheric exchange (STE) events and long-range transport from Eurasian biomass burning events contribute to the enhanced concentrations observed at Yellowstone National Park (Oltmans et al., 2010).

In addition to the high-elevation Yellowstone NP site, we investigated statistically significant coincidences at low-elevation monitoring sites across the US. We identified days of high probability for STT to enhance surface O_3 concentrations. One of the low-elevation monitoring sites investigated was located in Chittenden County, Vermont (394 m asl), The spring months of April 2006 (19 days), April 2008 (20 days) and May 2008 (17 days) exhibited enhanced O_3 concentrations (i.e., $\geq$ 50 ppb) that were statistically significantly related to STT-S events that reached the surface. Fig. 15 illustrates the frequency of the stratosphere-to-troposphere transport events to the surface at Yellowstone NP for the period. The most frequent occurrence of STT –S events were during the springtime months. There was no preference for the enhanced O_3 concentrations to occur during either the daylight or nighttime hours. During this period of time, both the daily maximum hourly average concentrations and the daily

maximum 8-h average concentrations were in the 40-60 ppb range. At the Chittenden County site in 2008, there was a major enhancement for the extended period 16-26 April. On 18 April, the maximum hourly average concentration was 89 ppb and the daily maximum 8-h concentration was 83 ppb. Coincidences between enhanced surface O_3 concentrations and STT-S events were indicated over the entire 11-day period. The daily maximum hourly average concentrations ranged from 57 to 89 ppb, with daily maximum 8-h averages from 55 to 83 ppb. There was evidence that the observed enhancement of the hourly average concentrations was affected by STT events.

At many of the other lower-elevation monitoring sites we investigated, numerous days occurred in which a strong relationship existed between enhanced O_3 concentrations and STT-S during the springtime. When statistically significant coincidences occurred at the lower elevation sites, the daily maximum hourly average concentrations were mostly in the 50-65 ppb range; the daily maximum 8-h average concentrations were usually in the 50-62 ppb range.

The US Environmental Protection Agency has indicated that O_3 background concentrations in the US are in the range of 15 to 35 ppb. Our results indicate that stratospheric-tropospheric exchange (STE) processes contribute to enhanced ($\geq$ 50 ppb) O_3 hourly averaged concentrations at the surface at both high and low-elevation monitoring sites. For the sites analyzed, results indicate that the coincidence between the number of days with a daily maximum hourly average O_3 concentration $\geq$ 50 ppb and STT-S >0 were frequent and exhibited a preference for occurrences during the springtime versus other times of the year.

2.4 Background contribution at the surface in the western US is larger than current models indicate

Jaffe (2011) examined O_3 data from 11 rural CASTNET sites in the western US for the period 1995-2009. The 11 surface sites show a similar seasonal cycle and generally a good correlation in the deseasonalized monthly means, indicating that there are large scale influences on O_3 that operate across the entire western US. These sites also show a good correlation between site elevation and annual mean O_3, indicating a significant contribution from the free troposphere. Jaffe (2011) examined the number of exceedance days for each site, defined as a day when the Maximum Daily 8-h Average (MDA8) exceeded a threshold value (i.e., 65 ppb, 70 ppb, or 75 ppb). Over this time period, more than half of these sites exceeded an MDA8 threshold of 70 ppb at least 4 times per year, and all sites exceeded a threshold value of 65 ppb at least 4 times per year. The transition to lower threshold values increased substantially the number of exceedance days, especially during spring, reflecting the fact that background O_3 peaks during spring. In addition, the author examined the correlation between surface O_3 and free tropospheric O_3 in the same region, as measured by routine balloon launches from Boulder, CO. Using O_3 measured by the balloon sensor in the range of 3-6 km above sea level, Jaffe (2011) reported statistically significant correlations between surface and free tropospheric O_3 in spring and summer months using both monthly means, daily MDA8 values, and the number of surface exceedance days. Jaffe (2011) suggested that during spring this correlation reflects variations in the flux of O_3 transport from the free troposphere to the surface. In summer, free tropospheric and surface concentrations of O_3 and the number of exceedance days were all significantly correlated with emissions from biomass burning in the western US. Jaffe (2011) noted that the correlation between surface and free tropospheric O_3 was strongest at lower exceedance thresholds, which suggested according to the author that a greater importance of background O_3

at the lower thresholds. Jaffe (2011) concluded that the large interannual variations and the correlation between surface and free tropospheric O_3 observed in his study during both spring and summer indicate that background O_3 varies strongly from year-to-year and that the background contribution at the surface in the western US is larger than current models indicate.

2.5 The application of PRB O_3 concentrations in risk assessments

As indicated previously, the EPA (2006; 2007) determined that PRB O_3 concentrations at the surface were generally in the range of 15 to 35 ppb. However, as indicated in the Introduction, the EPA used as input into its risk analysis estimates of PRB diurnal concentrations averaged on a monthly basis for the months of April through September. The PRB diurnal patterns were mostly in the 15 to 25 ppb range for the maximum monthly diurnal concentration (US EPA, 2007 – see Chapter 2 Appendix).

We have compared the PRB O_3 concentration predictions for the 12 cities that EPA focused on for its human health risk assessment with the concentrations measured at Trinidad Head, a site that has been used to provide an observational perspective on PRB O_3. The nighttime O_3 values at Trinidad Head are included for illustrative purposes only. The nighttime observations at Trinidad Head are often made under conditions that not representative of PRB. It is our opinion that the nighttime O_3 values observed at Trinidad Head are likely much lower than the O_3 concentrations that would be observed if PRB conditions prevailed. Figs. 16-21 illustrate the comparison between the predicted PRB diurnal patterns for the cities and the diurnal pattern of the hourly average concentrations for Trinidad Head for the months of April through September 2003. Note that for the months of April through June, which are part of the warm season months defined in the Staff Paper (US EPA, 2007), the diurnal pattern concentrations for

the diurnal patterns of hourly average PRB concentrations predicted for the 12 cities are less than the patterns derived from the empirical data at Trinidad Head. It is not until July that the diurnal PRB pattern for Trinidad Head is equivalent to the PRB hourly average concentrations predicted by the GEOS-Chem model for the 12 cities. For the months of August and September, the diurnal PRB pattern for Trinidad Head begins to rise above the GEOS-Chem model for the 12 cities. With few exceptions, the diurnal monthly PRB concentrations predicted by EPA are mostly in the 15 to 25 ppb range. Unlike the predicted hourly average PRB O_3 concentrations using the GEOS-Chem model, actual ***diurnal*** concentrations experienced during PRB conditions range from approximately 30 to 50 ppb. Measured ***hourly*** average PRB O_3 concentrations ≥ 50 ppb frequently occurred with maximum hourly average concentrations up to 65 ppb (Oltmans et al., 2008, 2010). The predicted PRB concentrations using models are below the observed PRB levels that influence the low-elevation monitoring site at Trinidad Head (CA) (Oltmans et al., 2008, 2010).

Hourly average concentrations frequently ≥ 50 ppb that are transported over the Pacific enter the west coast of the US and are transported eastward. Processes associated with PRB conditions replenish O_3 concentrations as O_3 is transported across the US (Jaffe et al., 2004; Bertschi et al., 2004; Bertschi and Jaffe, 2005; Pfister et al., 2010; Oltmans et al., 2010; Jaffe, 2011; Lefohn et al., 2011). Frequently occurring STT events during the springtime that result in enhanced hourly average concentrations ≥ 50 ppb at both high- and low-elevation monitoring sites and annually occurring wildfires in the US enhance PRB O_3 concentrations levels. GEOS-Chem models underestimate the frequently occurring hourly average PRB O_3 concentrations ≥ 50 ppb experienced during the springtime at the west coast of the US. In addition, the models do not adequately capture the replenishment processes associated with STT events that reach the

surface at both high- and low-elevation monitoring sites and the wildfires that enhance PRB O_3 hourly average concentrations.

3. Comments on Section 3.4 (Policy-Relevant Background) in ISA Ozone

Throughout the PRB section, the EPA has mostly used the multi-monthly (i.e., April-May or June-August) mean of the daily 8-h maximum concentration. By use the method of calculation, one is unable to accurately judge the performance of models when compared to the variability of the empirical data. PRB O_3 concentrations should not be characterized as a single average value, but rather all dimensions of temporal and spatial variability in PRB should be recognized. The characterization of model output when compared to empirical data should reflect the way in which the data are to be applied. For example, as indicated in the Introduction, the EPA during the past review cycle for the O_3 standard, used hourly averaged diurnal pattern concentrations as input into its risk assessment models.

For assessing the performance of the GEOS-Chem modeling results, the focus in Section 3.4 is on the mean daily 8-h maximum O_3 concentration over a multi-month period. The use of either a 2- or 3-month average concentration tends to not adequately characterize the variability associated with empirical data. For example, at the high-elevation monitoring site at Yellowstone NP in 2001, the April-May mean value for PRB O_3 was estimated to be 42.3 ± 5.6 ppb (page 3-38). This information tells us nothing about the range of hourly average concentrations associated with the site. In 2001, Yellowstone NP experienced 461 (April) and 350 (May) hourly average concentrations ≥ 50 ppb. In other words, over 800 hourly average O_3 concentrations ≥ 50 ppb occurred during the April-May period. Based on our analysis of STT events, we would estimate that many of these enhanced hourly average concentrations (i.e., hourly average

concentrations ≥ 50 ppb) experienced at Yellowstone NP were associated with PRB-related events. Similarly, on page 3-34, GEOS-Chem estimates that the mean value for PRB O_3 for April-May in the vicinity of the low-elevation site at Trinidad Head for 2001 is below 35 ppb. Generally at Trinidad Head, the mean value is approximately 45 ppb for April and May, with the number of hourly average concentrations ≥ 50 ppb for April varying over the range of 30 to 187 for the period 2002 to 2010 (see Table 1). For the Vermont Area, GEOS-Chem appears to predict for the April-May period a mean value for PRB O_3 below 35 ppb (page 3-34). As described earlier, Lefohn et al. (2011) reported at the low-elevation Chittenden County Vermont site a statistically significant relationship between STT-S and enhanced hourly average concentrations ≥ 50 ppb for April 2006 (19 days), April 2008 (20 days), and May 2008 (17 days). The month mean of the daily maximum 8-h average concentrations for these periods was 49 ppb, 54 ppb, and 46 ppb, respectively. In other words, for the months in which PRB conditions appears to contribute substantially to enhanced hourly average concentrations (i.e.,≥ 50 ppb), the monthly mean of the daily maximum 8-h average concentration was greater than the GEOS-Chem model predictions. The number of hourly average concentrations ≥ 50 ppb for April 2006, April 2008, and May 2008 was 134, 322, and 132, respectively. The 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.

Section 3.8.1 contains the time series plots of 8-h daily max O_3 concentrations observed at 14 CASTNET sites during 2001 and corresponding GEOS-Chem predictions for the base model (i.e., model including all anthropogenic and natural sources) and the PRB model (i.e., model including natural sources everywhere in the world and anthropogenic sources outside the U.S., Canada, and Mexico). Note that in Fig. 3A-9 for Yellowstone NP, the model does not appear to match the variability in the time series for the 8-h daily maximum concentration when compared to the actual measured values. Similar

observations occur for other CASTNET sites. The time series for the various sites appear to show a pattern of lower estimation of 8-h daily maximum concentrations than actual values at times during the springtime (e.g., Acadia NP, Crockett, Coffeeville, Voyageurs NP, Rocky Mountain NP, Yellowstone NP, Centennial, Canyonland NP, Mount Rainier NP, Lassen Volcanic NP, and Pinnacles NM). The lack of agreement may be associated with the model's inability to capture the O_3 concentration enhancements due to PRB conditions.

Although the EPA has attempted to summarize the science of PRB O_3, it is our opinion that further work is required to adequately assess the importance of the various PRB processes and their affects on defining PRB O_3 hourly average concentrations. Important contributions, although recognized in previous versions of the Criteria Document, such as the important contribution by Ludwig et al. (1977) are omitted. The work by Ludwig et al. (1977) was described in the Introduction. Evidence exists that STT processes are affecting springtime surface O_3 concentrations at both high- and low-elevation monitoring sites and the enhancements associated with the STT process have an important effect on estimating hourly average PRB O_3 concentrations.

4. Conclusion

In 2007, the EPA used estimates of PRB O_3 concentrations in the exposure and risk assessments for 12 urban areas in the US. The PRB O_3 concentrations used in the exposure and risk assessments were based on monthly diurnal patterns of hourly average concentration predictions of the model described in Fiore et al. (2003) for the 2001 April to September season. Correctly estimating hourly averaged PRB O_3 concentrations is very important because characterizing PRB O_3 levels that are too low can result in 1) overestimated human health risk

predictions and 2) optimistic policy expectations of the levels to which hourly average O_3 concentrations can be lowered as a result of emission reduction requirements.

Inaccurate statements concerning Trinidad Head are presented in Section 3.4 as to the usefulness of the O_3 observations to provide information on O_3 levels under PRB conditions. Meteorological evidence exists, and is presented here, to support the observation that conditions representative of PRB are routinely encountered at the low-elevation monitoring site at Trinidad Head, California. Trinidad Head regularly makes measurements under PRB 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 O_3 concentrations measured at this site. 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 PRB conditions. 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 (i.e., 30 to 187 hours). The range of maximum hourly average concentrations for the month of April over the period 2002-2010 is 54 to 65 ppb. Generally, during April and May the monthly mean of the daily maximum 8-h average concentration is approximately 45 ppb, with the number of hourly average concentrations ≥ 50 ppb for April varying over the range of 30 to 187 for the period 2002 to 2010.

Eurasian emissions associated with biomass burning and their easterly transport contribute to O_3 concentrations at west coast O_3 monitoring sites under PRB conditions, as well as inland O_3 monitoring sites in Montana, Wyoming, and North Dakota. Published information indicates that biomass burning events in Eurasia are not infrequent. Naturally occurring stratospheric-tropospheric exchange (STE) processes contribute to background O_3 concentrations

at the surface at both high and low-elevation monitoring sites. Natural processes, such as STE and biogenic emissions, contribute to the replenishment of PRB O_3 concentrations across the entire United States. Indications at many of the lower-elevation monitoring sites investigated using the LAGRANTO model are that numerous days occur in which a strong relationship exist between enhanced O_3 concentrations and stratosphere-to-troposphere transport to surface (STT-S) during the springtime. When statistically significant coincidences occurred at the lower elevation sites, the daily maximum hourly average concentrations were mostly in the 50-65 ppb range; the daily maximum 8-h average concentrations were usually in the 50-62 ppb range.

Evidence exists that show that background O_3 varies strongly from year-to-year and that the background contribution at the surface in the western US is larger are larger than current models indicate. GEOS-Chem models underestimate the frequently occurring hourly average PRB O_3 concentrations $\geq$ 50 ppb experienced during the springtime at the west coast of the US. In addition, the models do not adequately capture the replenishment processes associated with STT events that reach the surface at both high- and low-elevation monitoring sites and the wildfires that enhance PRB O_3 hourly average concentrations.

Throughout Section 3.4, the EPA has mostly used the multi-monthly (i.e., April-May or June-August) mean of the daily 8-h maximum concentration. By using the method of calculation, one is unable to accurately judge the performance of models when compared to the variability of the empirical data. The characterization of model output when compared to empirical data should reflect the way in which the data are to be applied. PRB O_3 concentrations should not be characterized as a single average value, but rather all dimensions of temporal and spatial variability in PRB should be recognized. For example, in the last review cycle for the O_3

standard, the EPA used hourly averaged diurnal pattern concentrations as input into its risk assessment models.

The 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. For example, at the high-elevation monitoring site at Yellowstone NP in 2001, the April-May mean value for PRB O_3 was estimated to be 42.3 ± 5.6 ppb (page 3-38). This information tells us nothing about the range of hourly average concentrations associated with the site. In 2001, Over 800 hourly average O_3 concentrations ≥ 50 ppb occurred during the April-May period. Based on our analysis of STT events, we would estimate that many of these enhanced hourly average concentrations (i.e., hourly average concentrations ≥ 50 ppb) experienced at Yellowstone NP were associated with PRB-related events. Similarly, on page 3-34, GEOS-Chem estimates that the mean value for PRB O_3 for April-May in the vicinity of the low-elevation site at Trinidad Head for 2001 is below 35 ppb. Generally at Trinidad Head, the mean value is approximately 45 ppb for April and May, with the number of hourly average concentrations ≥ 50 ppb for April varying over the range of 30 to 187 for the period 2002 to 2010. For the Vermont Area, GEOS-Chem appears to predict for the April-May period a mean value for PRB O_3 below 35 ppb (page 3-34). For the low-elevation Chittenden County Vermont site, we report a statistically significant relationship between STT-S and enhanced hourly average concentrations ≥ 50 ppb for April 2006 (19 days), April 2008 (20 days), and May 2008 (17 days). The month mean of the daily maximum 8-h average concentrations for these periods was 49 ppb, 54 ppb, and 46 ppb, respectively. In other words, for the months in which PRB conditions appears to contribute substantially to enhanced hourly average concentrations (i.e.,≥ 50 ppb), the monthly mean of the daily maximum 8-h average

concentration was greater than the GEOS-Chem model predictions. The number of hourly average concentrations ≥ 50 ppb for April 2006, April 2008, and May 2008 was 134, 322, and 132, respectively.

Section 3.8.1 contains the time series plots of 8-h daily max O_3 concentrations observed at 14 CASTNET sites during 2001 and corresponding GEOS-Chem predictions for the base model (i.e., model including all anthropogenic and natural sources) and the PRB model (i.e., model including natural sources everywhere in the world and anthropogenic sources outside the U.S., Canada, and Mexico). The time series for the various sites appear to show a pattern of lower estimation of 8-h daily maximum concentrations than actual values at times during the springtime (e.g., Acadia NP, Crockett, Coffeeville, Voyageurs NP, Rocky Mountain NP, Yellowstone NP, Centennial, Canyonland NP, Mount Rainier NP, Lassen Volcanic NP, and Pinnacles NM). The lack of agreement may be associated with the model's inability to capture the O_3 concentration enhancements due to PRB conditions.

Although the EPA has attempted to summarize the science of PRB O_3, it is our opinion that further discussion is required in the document to adequately assess the importance of the various PRB processes and their affects on defining PRB O_3 hourly average concentrations. Important contributions, although cited in previous versions of the Criteria Document, such as the important contribution by Ludwig et al. (1977), are omitted in the current version. Evidence exists that frequently occurring STT processes are affecting springtime surface O_3 concentrations at both high- and low-elevation monitoring sites and the enhancements associated with the STT process have an important effect on estimating hourly average PRB O_3 concentrations.

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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 9-year period, 2002 through 2010. 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)

Table 2. Summary for Trinidad Head, CA, of hourly average percentiles (ppm), number of hourly occurrences ≥ 50 ppb (N50), 24-hr SUM06 (ppm-hr), and 24-hr W126 (ppm-hr) cumulative exposure values for 2003-2010. UTC time period.

Year	Month	N	Min	P10	P30	P50	P70	P90	P95	P99	Max	N50	SUM06	W126
2003	January	739	0.008	0.014	0.021	0.028	0.034	0.038	0.041	0.044	0.046	0	0.000	0.312
2003	February	660	0.014	0.023	0.029	0.034	0.038	0.044	0.045	0.048	0.050	1	0.000	0.587
2003	March	713	0.012	0.028	0.034	0.038	0.042	0.047	0.049	0.051	0.052	23	0.000	1.076
2003	April	720	0.023	0.036	0.042	0.046	0.048	0.052	0.054	0.058	0.064	187	0.310	2.722
2003	May	743	0.011	0.029	0.036	0.041	0.045	0.050	0.052	0.054	0.057	96	0.000	1.773
2003	June	720	0.016	0.025	0.032	0.037	0.040	0.044	0.046	0.053	0.054	13	0.000	0.816
2003	July	744	0.003	0.015	0.020	0.023	0.026	0.031	0.032	0.036	0.041	0	0.000	0.109
2003	August	740	0.007	0.020	0.024	0.027	0.030	0.034	0.035	0.038	0.041	0	0.000	0.185
2003	September	720	0.006	0.016	0.025	0.030	0.035	0.040	0.042	0.054	0.060	13	0.120	0.507
2003	October	743	0.004	0.016	0.025	0.028	0.033	0.038	0.040	0.044	0.047	0	0.000	0.300
2003	November	696	0.006	0.023	0.028	0.032	0.035	0.039	0.041	0.046	0.047	0	0.000	0.396
2003	December	725	0.004	0.024	0.029	0.033	0.037	0.041	0.042	0.045	0.050	1	0.000	0.492
2004	January	742	0.001	0.022	0.027	0.032	0.036	0.041	0.043	0.047	0.052	2	0.000	0.478
2004	February	691	0.014	0.027	0.033	0.037	0.041	0.045	0.047	0.051	0.052	13	0.000	0.882
2004	March	744	0.009	0.019	0.027	0.035	0.042	0.047	0.050	0.054	0.062	38	0.184	1.127
2004	April	719	0.011	0.026	0.035	0.040	0.044	0.048	0.049	0.052	0.054	30	0.000	1.303
2004	May	734	0.008	0.016	0.031	0.035	0.039	0.043	0.045	0.048	0.050	3	0.000	0.691
2004	June	719	0.013	0.023	0.029	0.032	0.035	0.040	0.043	0.047	0.050	2	0.000	0.452
2004	July	744	0.004	0.014	0.019	0.023	0.026	0.031	0.033	0.036	0.038	0	0.000	0.104
2004	August	743	0.005	0.014	0.020	0.024	0.027	0.033	0.035	0.041	0.044	0	0.000	0.142
2004	September	697	0.010	0.020	0.025	0.029	0.033	0.037	0.039	0.042	0.044	0	0.000	0.272
2004	October	744	0.006	0.019	0.025	0.029	0.033	0.038	0.040	0.044	0.046	0	0.000	0.322
2004	November	719	0.001	0.011	0.020	0.025	0.029	0.035	0.036	0.041	0.043	0	0.000	0.169
2004	December	744	0.002	0.012	0.023	0.030	0.034	0.039	0.040	0.044	0.045	0	0.000	0.330
2005	January	744	0.007	0.017	0.025	0.030	0.035	0.039	0.042	0.044	0.047	0	0.000	0.380
2005	February	670	0.011	0.021	0.027	0.031	0.036	0.041	0.043	0.047	0.050	4	0.000	0.431
2005	March	700	0.006	0.020	0.032	0.039	0.045	0.049	0.051	0.053	0.055	58	0.000	1.377
2005	April	720	0.011	0.027	0.030	0.040	0.044	0.050	0.052	0.054	0.056	72	0.000	1.455
2005	May	632	0.011	0.024	0.030	0.034	0.037	0.043	0.046	0.050	0.052	6	0.000	0.561
2005	June	720	0.008	0.019	0.025	0.028	0.032	0.039	0.041	0.044	0.046	0	0.000	0.301
2005	July	744	0.005	0.015	0.020	0.022	0.027	0.031	0.033	0.036	0.039	0	0.000	0.111
2005	August	744	0.003	0.012	0.017	0.022	0.026	0.030	0.032	0.0335	0.038	0	0.000	0.096
2005	September	720	0.005	0.013	0.020	0.025	0.029	0.034	0.036	0.040	0.044	0	0.000	0.162
2005	October	744	0.003	0.014	0.020	0.025	0.031	0.037	0.039	0.042	0.047	0	0.000	0.221
2005	November	719	0.003	0.013	0.021	0.028	0.033	0.039	0.041	0.044	0.046	0	0.000	0.289
2005	December	737	0.006	0.017	0.022	0.028	0.034	0.040	0.042	0.049	0.050	4	0.000	0.386

Table 2 (Continued). Summary for Trinidad Head, CA, of hourly average percentiles (ppm), number of hourly occurrences ≥ 50 ppb (N50), 24-hr SUM06 (ppm-hr), and 24-hr W126 (ppm-hr) cumulative exposure values for 2003-2010. UTC time period.

Year	Month	N	Min	P10	P30	P50	P70	P90	P95	P99	Max	N50	SUM06	W126
2006	January	668	0.017	0.028	0.031	0.034	0.036	0.042	0.044	0.047	0.047	0	0.000	0.528
2006	February	671	0.010	0.024	0.030	0.036	0.040	0.044	0.045	0.049	0.054	5	0.000	0.690
2006	March	741	0.019	0.033	0.039	0.042	0.045	0.048	0.050	0.053	0.055	37	0.000	1.611
2006	April	720	0.012	0.030	0.036	0.041	0.044	0.049	0.051	0.056	0.060	56	0.060	1.510
2006	May	698	0.010	0.028	0.034	0.039	0.043	0.048	0.050	0.060	0.064	43	0.438	1.340
2006	June	720	0.006	0.019	0.026	0.030	0.033	0.037	0.039	0.043	0.047	0	0.000	0.298
2006	July	744	0.010	0.016	0.021	0.023	0.026	0.030	0.031	0.034	0.035	0	0.000	0.102
2006	August	715	0.007	0.017	0.022	0.026	0.030	0.032	0.034	0.037	0.039	0	0.000	0.149
2006	September	718	0.005	0.015	0.022	0.027	0.032	0.039	0.043	0.048	0.050	1	0.000	0.338
2006	October	744	0.005	0.017	0.026	0.030	0.034	0.039	0.041	0.048	0.051	2	0.000	0.379
2006	November	720	0.007	0.019	0.027	0.033	0.038	0.042	0.044	0.045	0.048	0	0.000	0.530
2006	December	685	0.010	0.020	0.025	0.030	0.035	0.041	0.042	0.045	0.048	0	0.000	0.373
2007	January	696	0.005	0.016	0.023	0.027	0.032	0.037	0.039	0.042	0.043	0	0.000	0.235
2007	February	671	0.008	0.023	0.031	0.035	0.039	0.044	0.045	0.047	0.048	0	0.000	0.645
2007	March	743	0.008	0.022	0.030	0.035	0.040	0.046	0.049	0.051	0.052	21	0.000	0.896
2007	April	719	0.006	0.027	0.034	0.039	0.043	0.048	0.050	0.056	0.057	46	0.000	1.336
2007	May	744	0.014	0.026	0.033	0.038	0.042	0.048	0.049	0.052	0.055	30	0.000	1.164
2007	June	720	0.013	0.021	0.025	0.028	0.032	0.035	0.037	0.039	0.040	0	0.000	0.214
2007	July	700	0.004	0.013	0.019	0.023	0.025	0.029	0.030	0.033	0.033	0	0.000	0.079
2007	August	743	0.006	0.016	0.021	0.024	0.027	0.031	0.033	0.039	0.043	0	0.000	0.125
2007	September	720	0.010	0.019	0.025	0.029	0.032	0.037	0.039	0.042	0.044	0	0.000	0.268
2007	October	742	0.006	0.017	0.023	0.028	0.032	0.038	0.040	0.045	0.048	0	0.000	0.294
2007	November	720	0.004	0.016	0.023	0.027	0.031	0.038	0.041	0.046	0.048	0	0.000	0.271
2007	December	569	0.004	0.019	0.027	0.032	0.035	0.040	0.041	0.043	0.043	0	0.000	0.308
2008	January	743	0.006	0.019	0.031	0.035	0.037	0.041	0.043	0.046	0.047	0	0.000	0.553
2008	February	696	0.009	0.015	0.023	0.033	0.037	0.043	0.045	0.049	0.051	4	0.000	0.539
2008	March	738	0.015	0.027	0.034	0.039	0.043	0.048	0.049	0.052	0.053	26	0.000	1.261
2008	April	703	0.011	0.029	0.036	0.042	0.047	0.052	0.055	0.057	0.061	120	0.061	1.989
2008	May	722	0.007	0.023	0.031	0.037	0.043	0.048	0.049	0.053	0.056	34	0.000	1.123
2008	June	697	0.012	0.021	0.026	0.031	0.035	0.040	0.042	0.045	0.052	3	0.000	0.398
2008	July	744	0.006	0.015	0.021	0.024	0.027	0.033	0.036	0.040	0.042	0	0.000	0.147
2008	August	744	0.003	0.013	0.018	0.021	0.024	0.030	0.031	0.034	0.035	0	0.000	0.079
2008	September	719	0.007	0.016	0.023	0.027	0.031	0.036	0.038	0.042	0.045	0	0.000	0.220
2008	October	714	0.005	0.015	0.022	0.027	0.032	0.037	0.039	0.044	0.052	2	0.000	0.257
2008	November	720	0.005	0.013	0.020	0.025	0.030	0.036	0.039	0.043	0.045	0	0.000	0.210
2008	December	744	0.006	0.016	0.026	0.030	0.034	0.038	0.040	0.043	0.044	0	0.000	0.322

Table 2 (Continued). Summary for Trinidad Head, CA, of hourly average percentiles (ppm), number of hourly occurrences ≥ 50 ppb (N50), 24-hr SUM06 (ppm-hr), and 24-hr W126 (ppm-hr) cumulative exposure values for 2003-2010. UTC time period.

Year	Month	N	Min	P10	P30	P50	P70	P90	P95	P99	Max	N50	SUM06	W126
2009	January	741	0.006	0.017	0.022	0.026	0.030	0.036	0.038	0.042	0.044	0	0.000	0.209
2009	February	672	0.011	0.024	0.033	0.037	0.040	0.043	0.046	0.048	0.051	2	0.000	0.705
2009	March	720	0.014	0.028	0.035	0.039	0.043	0.047	0.048	0.050	0.054	18	0.000	1.148
2009	April	720	0.011	0.026	0.035	0.041	0.045	0.050	0.052	0.061	0.065	73	0.684	1.763
2009	May	718	0.011	0.021	0.029	0.034	0.038	0.043	0.045	0.051	0.054	11	0.000	0.656
2009	June	720	0.011	0.021	0.026	0.028	0.030	0.033	0.034	0.038	0.043	0	0.000	0.183
2009	July	744	0.006	0.014	0.020	0.022	0.025	0.030	0.031	0.033	0.035	0	0.000	0.088
2009	August	713	0.005	0.013	0.019	0.023	0.026	0.030	0.031	0.036	0.038	0	0.000	0.093
2009	September	718	0.006	0.016	0.021	0.026	0.030	0.034	0.036	0.039	0.042	0	0.000	0.170
2009	October	744	0.004	0.012	0.021	0.026	0.031	0.036	0.039	0.042	0.044	0	0.000	0.222
2009	November	720	0.004	0.015	0.025	0.031	0.036	0.040	0.042	0.044	0.046	0	0.000	0.393
2009	December	743	0.006	0.016	0.022	0.028	0.032	0.038	0.040	0.042	0.045	0	0.000	0.277
2010	January	741	0.005	0.018	0.026	0.032	0.037	0.043	0.044	0.047	0.049	0	0.000	0.558
2010	February	666	0.006	0.019	0.028	0.033	0.036	0.041	0.044	0.047	0.051	3	0.000	0.463
2010	March	744	0.012	0.025	0.033	0.039	0.043	0.047	0.050	0.053	0.058	38	0.000	1.285
2010	April	703	0.014	0.030	0.037	0.042	0.045	0.049	0.050	0.054	0.058	51	0.000	1.510
2010	May	744	0.013	0.028	0.033	0.037	0.040	0.044	0.045	0.049	0.052	6	0.000	0.825
2010	June	720	0.008	0.020	0.024	0.028	0.031	0.035	0.036	0.040	0.042	0	0.000	0.203
2010	July	744	0.005	0.015	0.019	0.023	0.025	0.028	0.029	0.031	0.032	0	0.000	0.078
2010	August	737	0.005	0.015	0.019	0.022	0.025	0.030	0.033	0.036	0.038	0	0.000	0.096
2010	September	698	0.003	0.009	0.014	0.020	0.025	0.032	0.034	0.036	0.041	0	0.000	0.091
2010	October	720	0.008	0.020	0.027	0.031	0.034	0.038	0.041	0.043	0.048	0	0.000	0.347
2010	November	720	0.005	0.015	0.024	0.030	0.034	0.039	0.042	0.045	0.046	0	0.000	0.340
2010	December	744	0.013	0.024	0.031	0.035	0.038	0.042	0.044	0.046	0.049	0	0.000	0.642

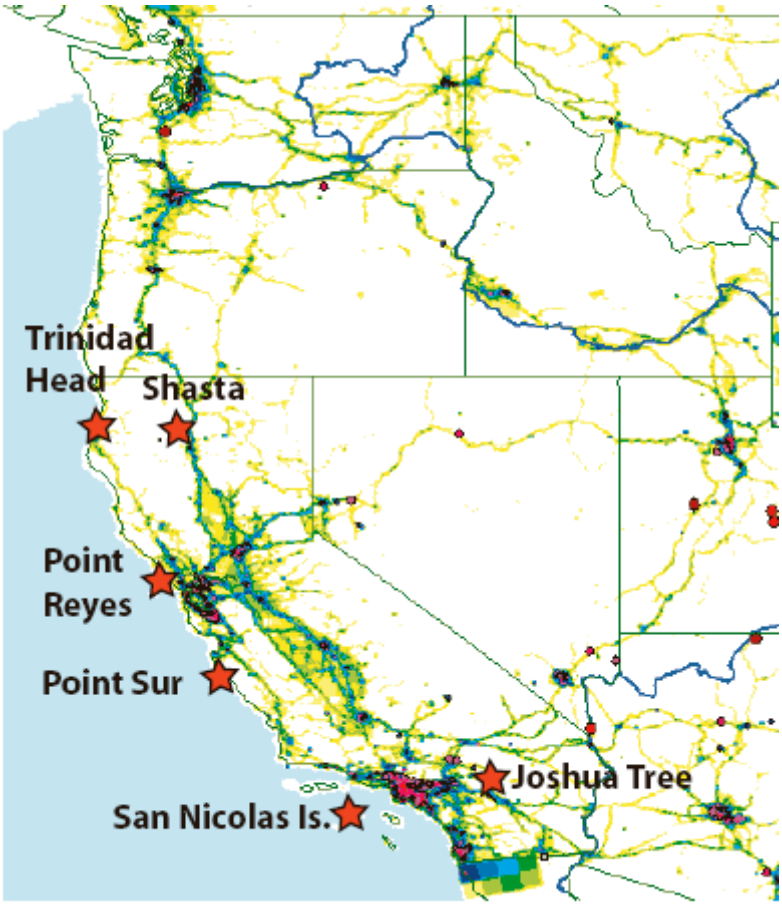

Fig. 1. Map of the location of the surface ozone and ozonesonde measurement site at Trinidad Head and ozonesonde sites in California during the IONS 2010/CalNex campaign.

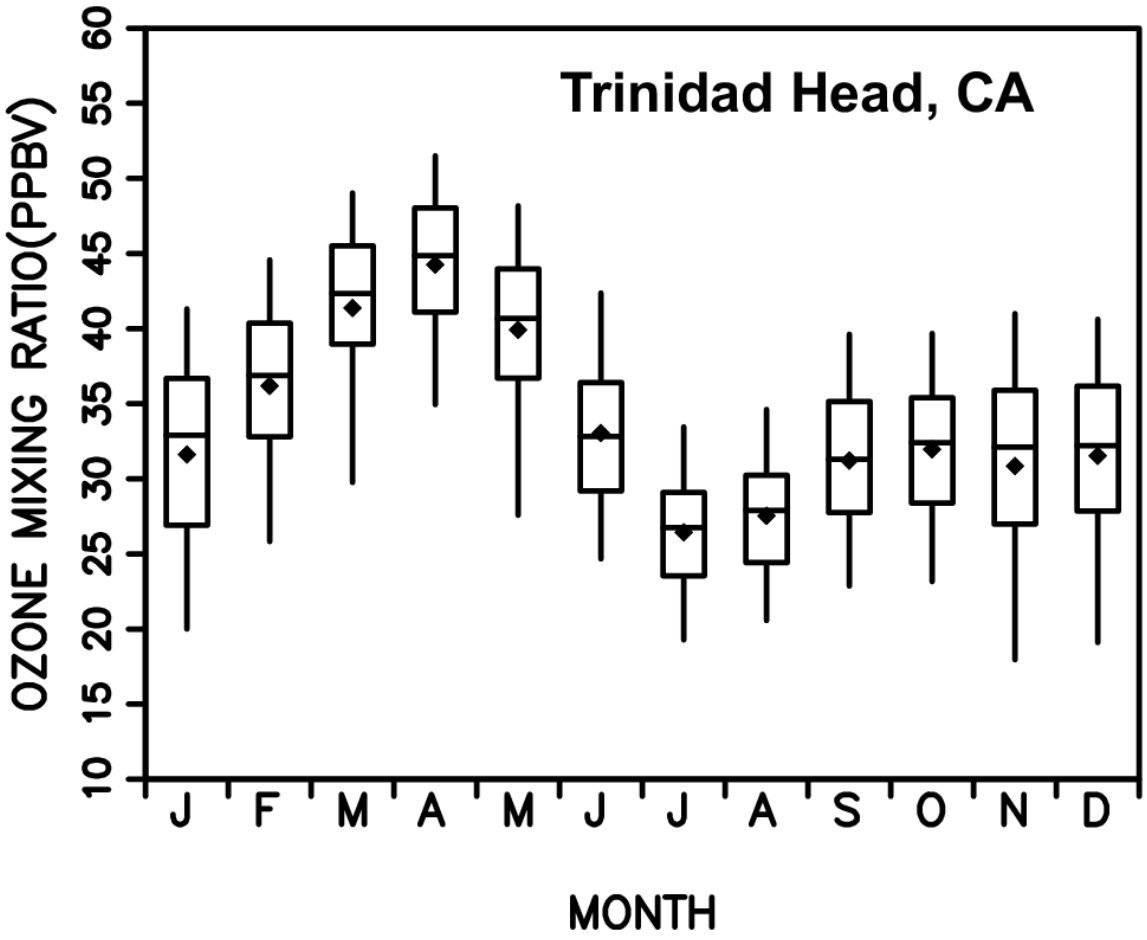

Fig. 2. Seasonal variation of daytime (1100-1800 Local Standard Time) surface ozone at Trinidad Head, California (41N). The diamond is the mean, the horizontal bar within the box is the median, the box is the inner 50th percentile and the whiskers are the inner 90th percentile of the hourly averages based on data from 2002-2010.

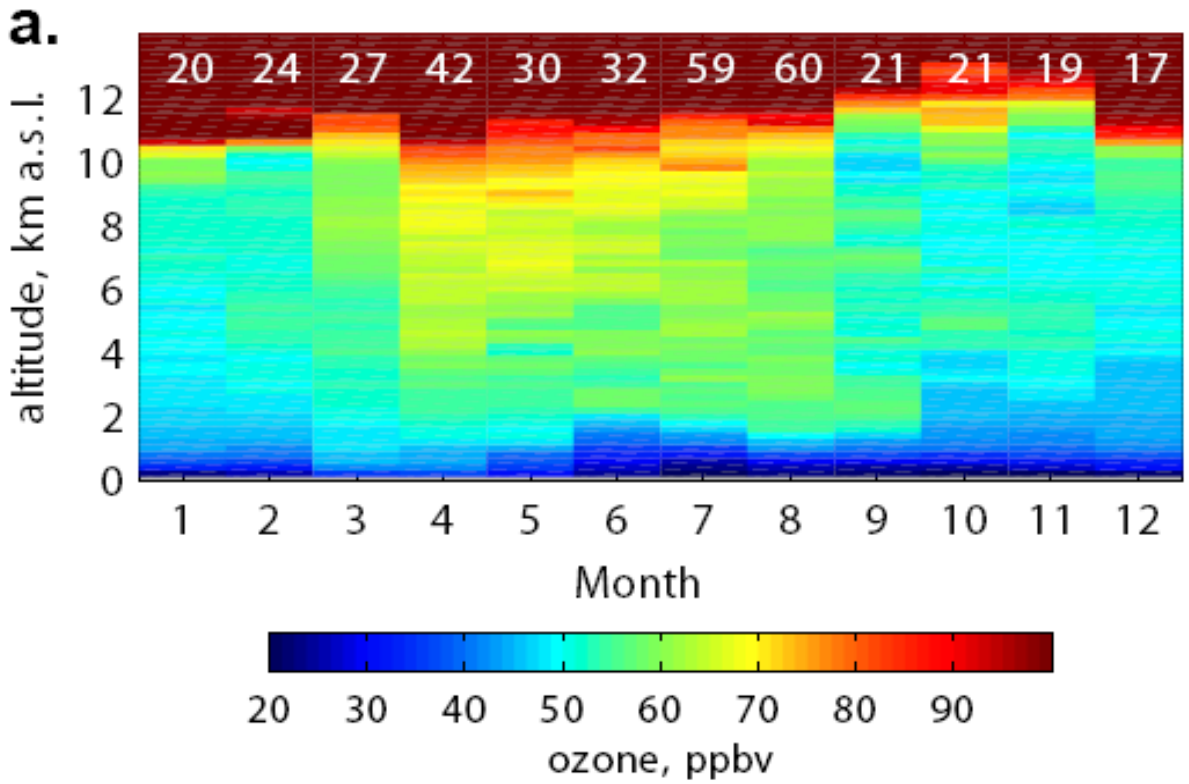

Fig. 3. Tropospheric ozone mixing ratios (ppbv) above the Trinidad Head Observatory from ozonsonde measurements. The numbers at the top of the figure for each month are the number of soundings for that month in the period 2004-2010 (from Cooper et al., 2011).

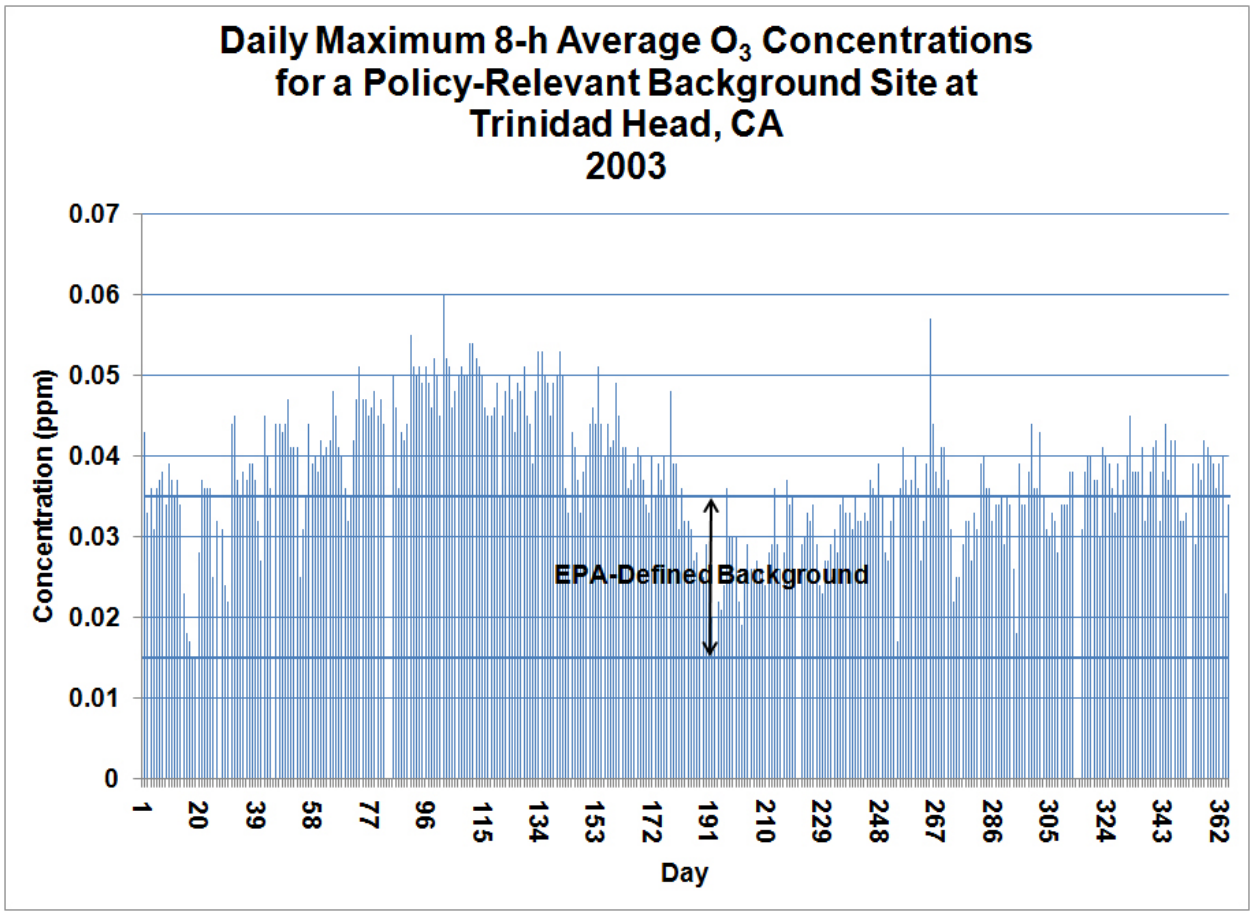

Fig. 4. Daily maximum 8-h average ozone concentrations Trinidad Head, CA for 2003. The range of the EPA defined policy-relevant background ozone concentrations of 15 to 35 ppb, based on modeling, is identified.

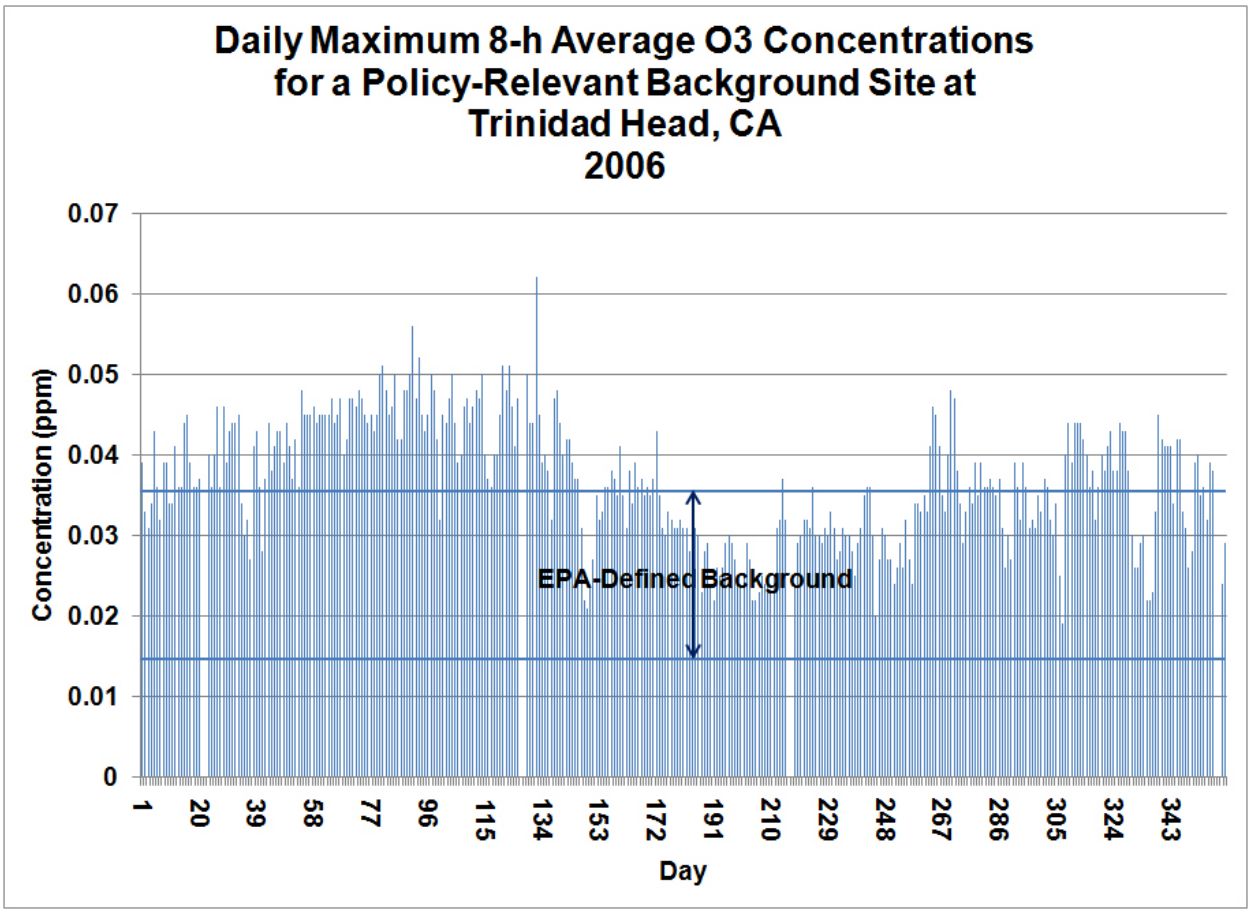

Fig. 5. Daily maximum 8-h average ozone concentrations Trinidad Head, CA for 2006. The range of the EPA defined policy-relevant background ozone concentrations of 15 to 35 ppb, based on modeling, is identified.

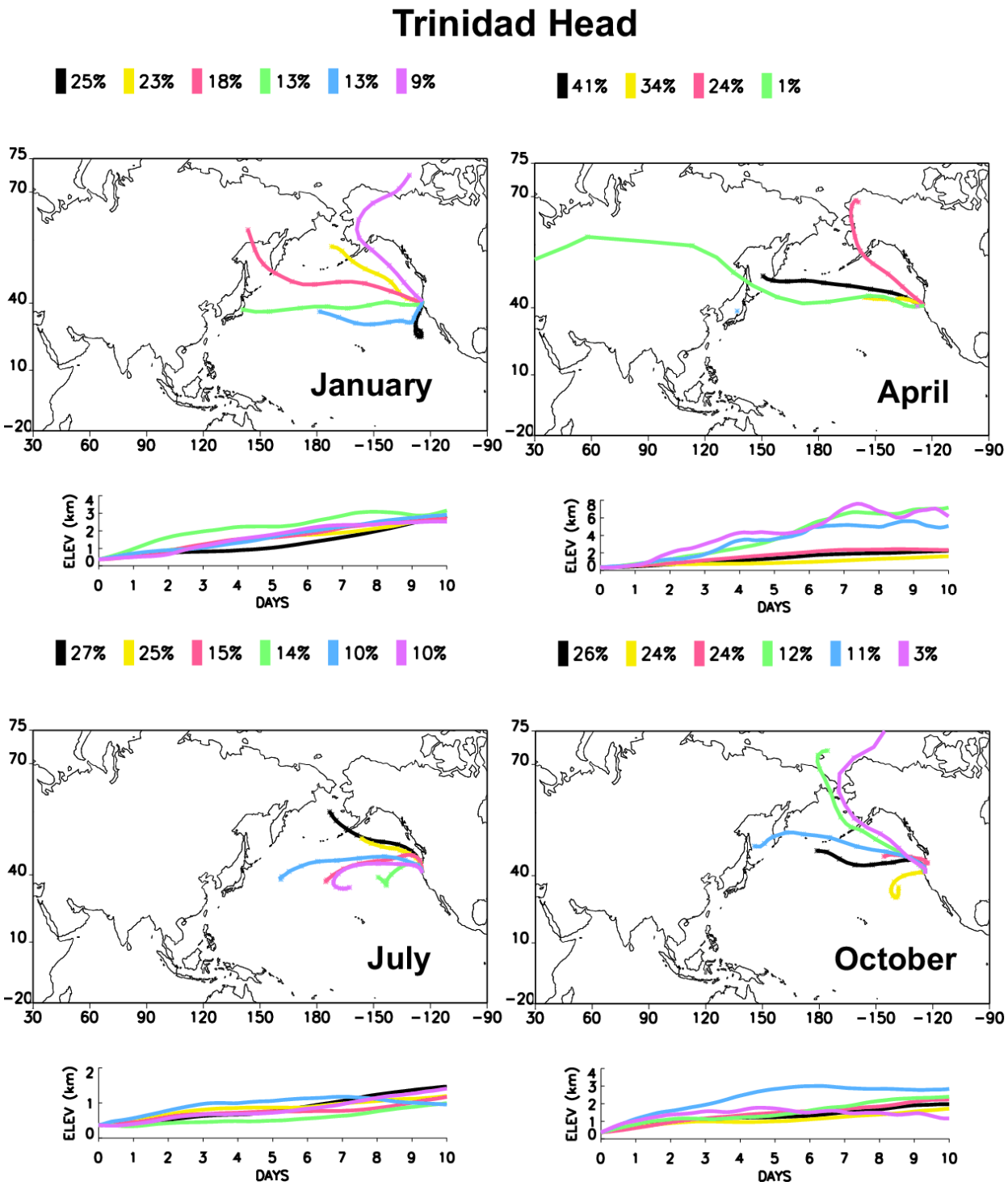

Fig. 6. Trajectory clusters for Trinidad Head, California for years 2002 to 2005 showing average flow characteristics at the station (from Oltmans et al., 2008).

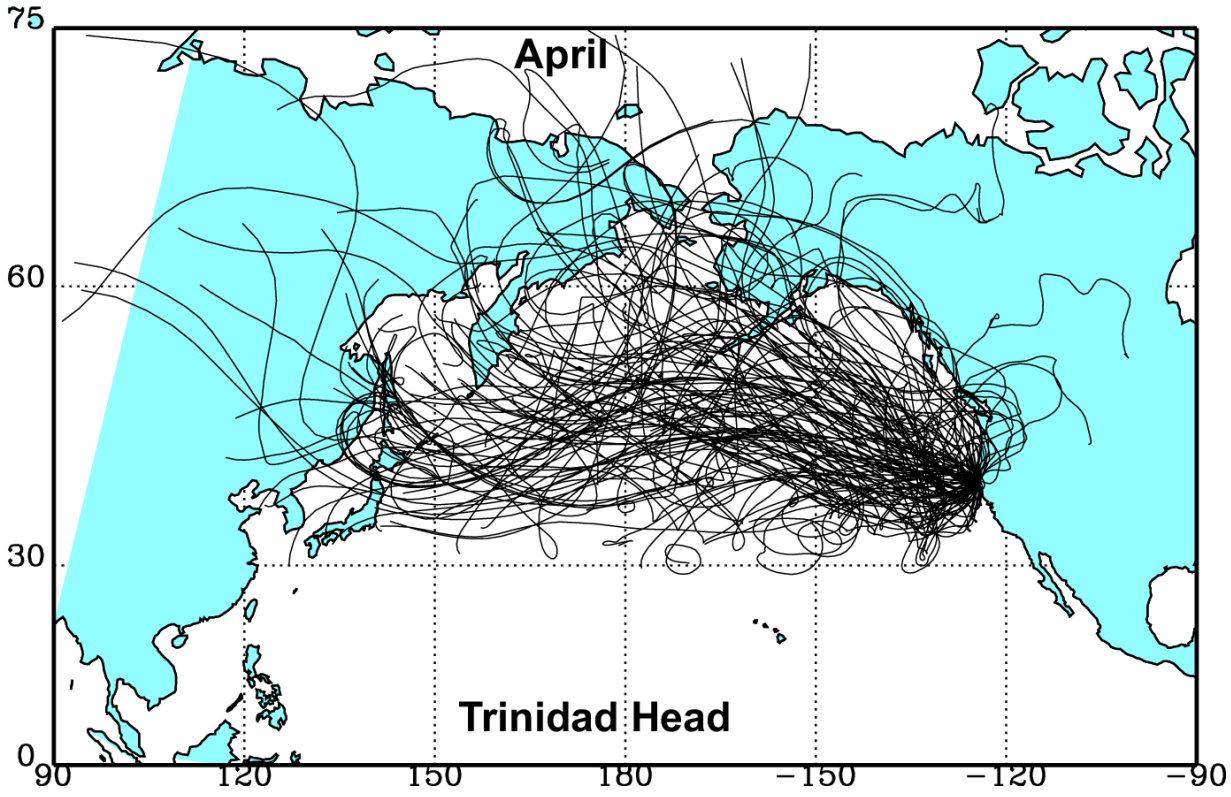

Fig. 7. Back trajectories from Trinidad Head, California on days with hourly average O_3 amounts ≥ 50 ppb for April in the years 2002 to 2005 (from Oltmans et al., 2008).

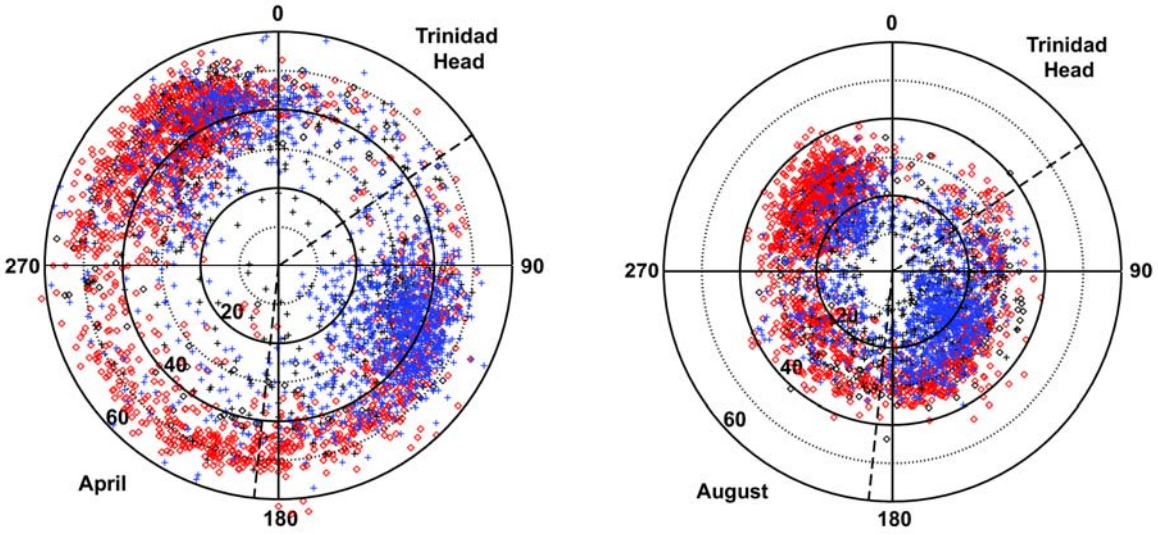

Fig. 8. Wind rose for ozone mixing ratios at Trinidad Head for April and August for the period 2002-2007. The concentric circles mark the ozone mixing from 0 ppbv at the center to 60 ppbv for the outer circle. The red diamonds are daytime (10-21 LST) values and the blue plusses are nighttime (22-09 LST) 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 (56^0 to 186^0), the wind is blowing from over the land to the observing site (from Oltmans et al., 2008).

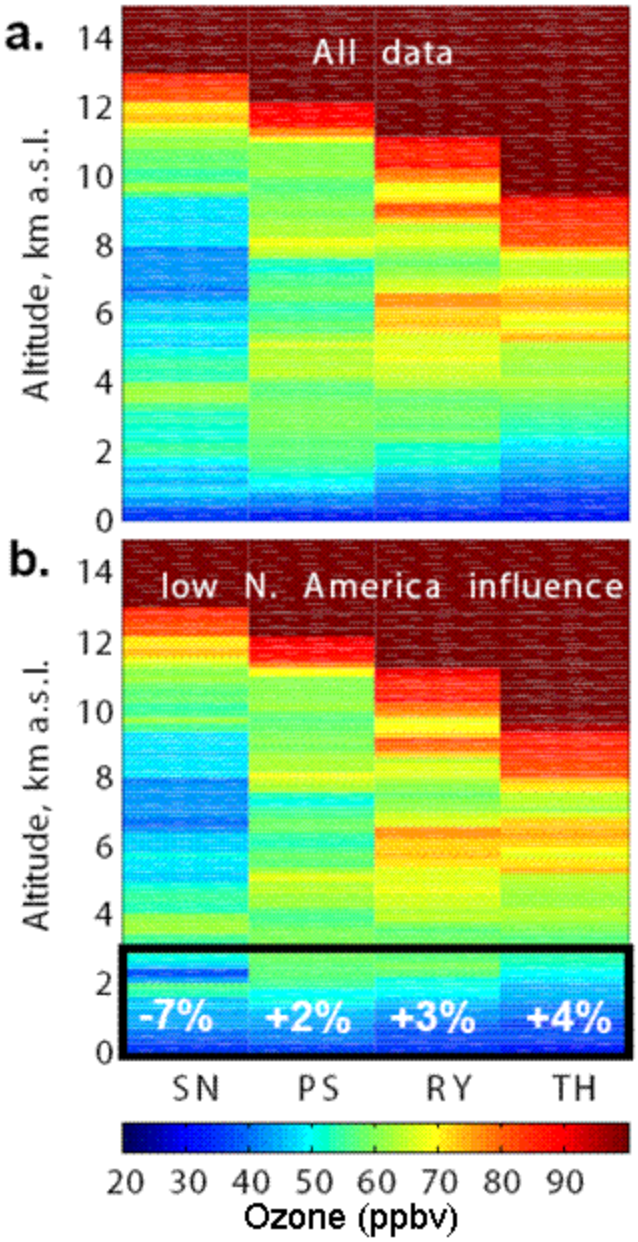

Fig. 9. a.) Median ozone profiles above the four coastal sites using all available data; b.) Same as in a.) but measurements with recent North American anthropogenic influence removed. Numbers in white indicate the percent change in the mass of ozone in the 0-3 km range when air masses with strong North American influence are removed. None of these changes are statistically significant (from Cooper et al., 2011).

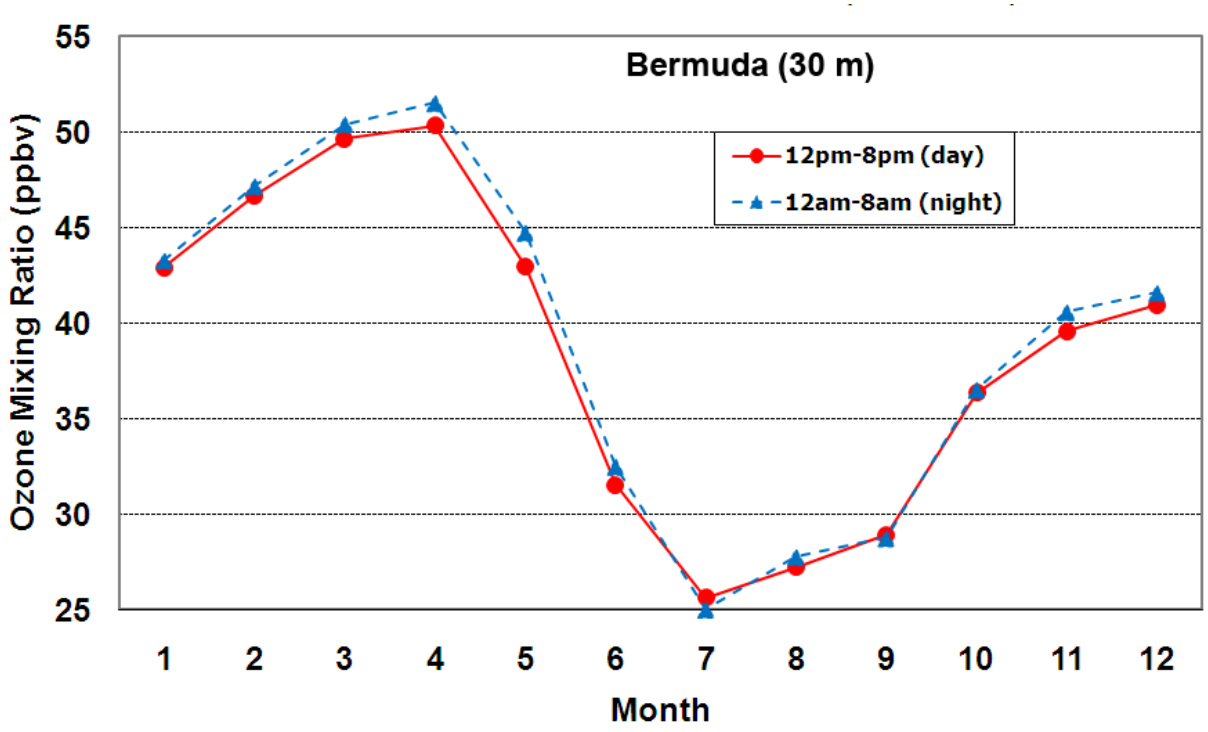

Fig. 10. Monthly ozone levels at Bermuda for day (red) and night (blue) for each month demonstrating that at a relatively remote site with minimal influence from local circulation effects there is essentially no difference between ozone levels during the day and night.

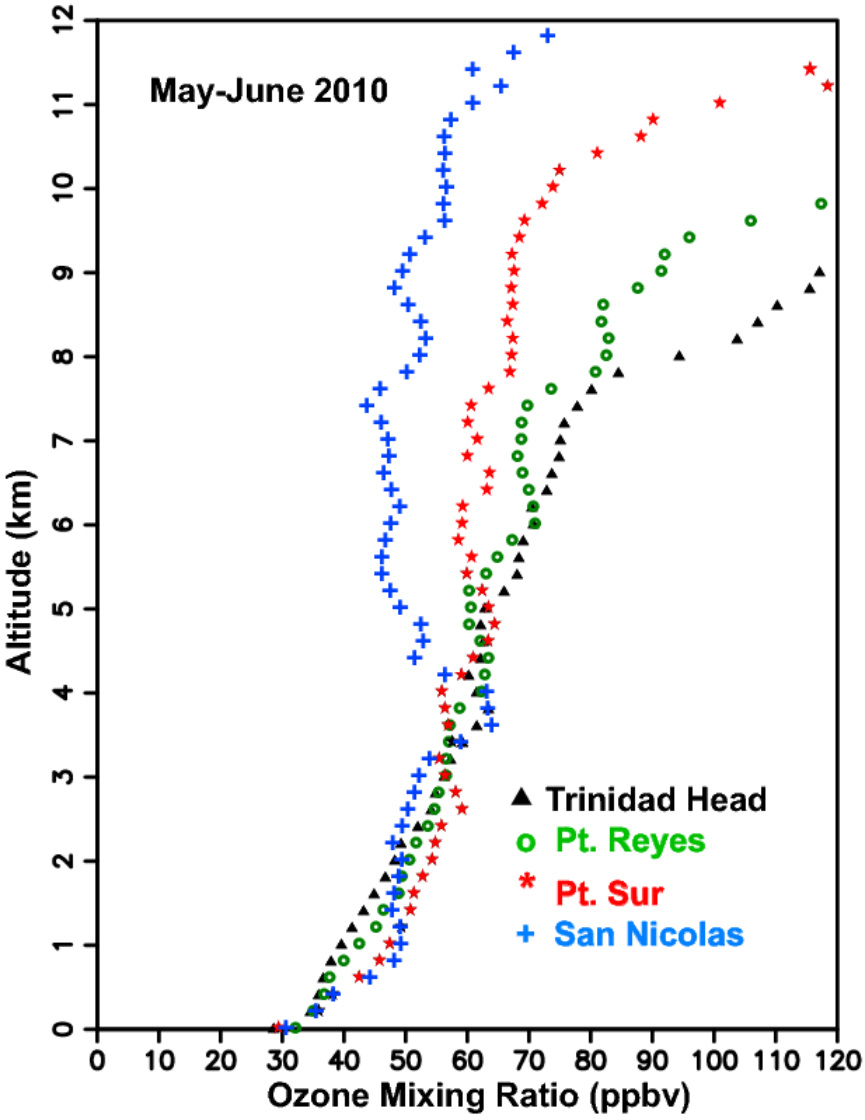

Fig. 11. Average ozone mixing ratio profiles at four sites making ozonesonde observations during the IONS 2010/CalNex Campaign in May-June 2010. In the lower troposphere (<4 km) average ozone amounts are similar along the California coast.

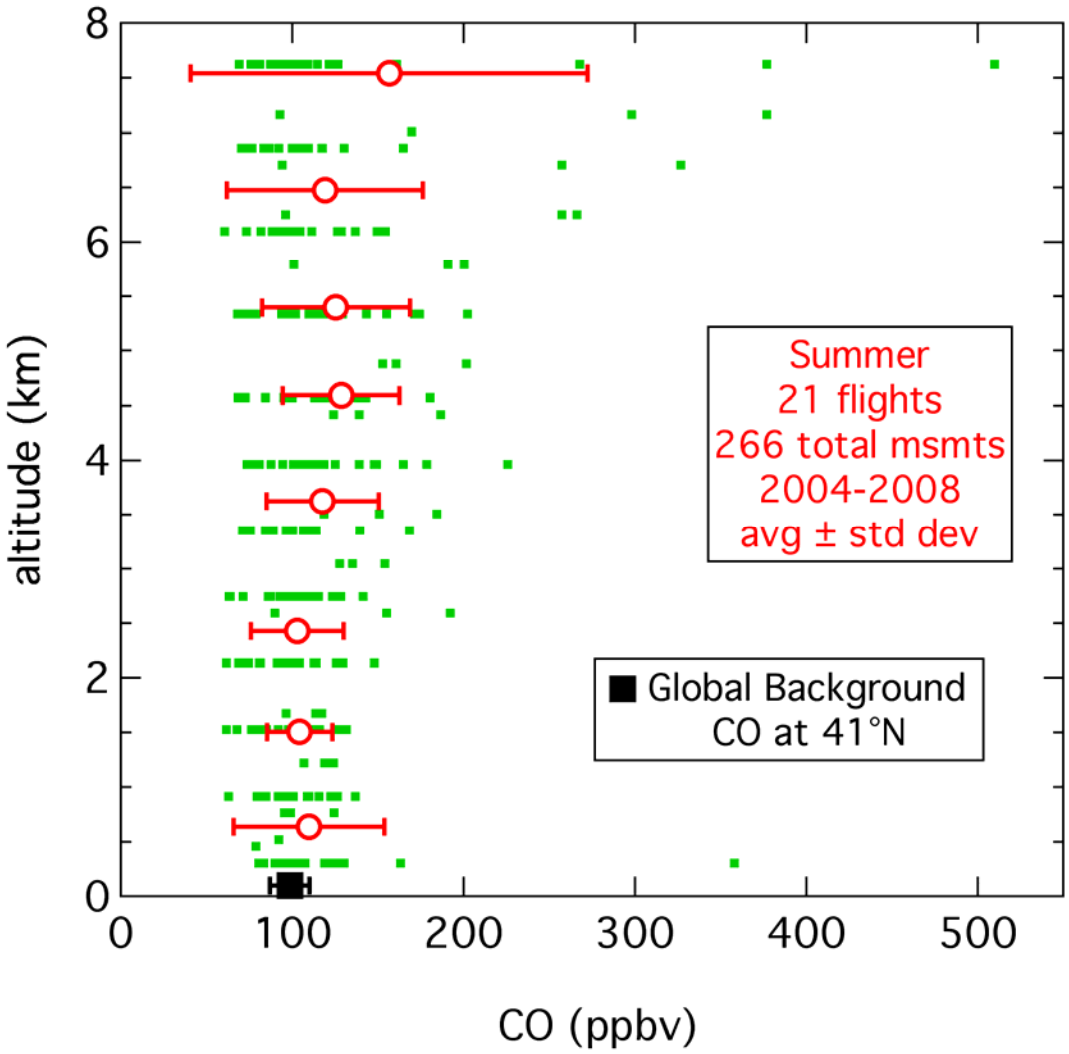

Fig. 12. Vertical profiles of carbon monoxide measured in flasks collected on aircraft flights above Trinidad Head CA. The green points give the individual measurements, and the red circles indicate averages and standard deviations for 1 km altitude segments. The black symbol gives the surface global carbon monoxide background as determined from the NOAA ESRL Global Monitoring Division flask network for the years of the aircraft measurements (Paul Novelli, NOAA ESRL Global Monitoring Division, private communication.) (from Parrish et al., 2010).

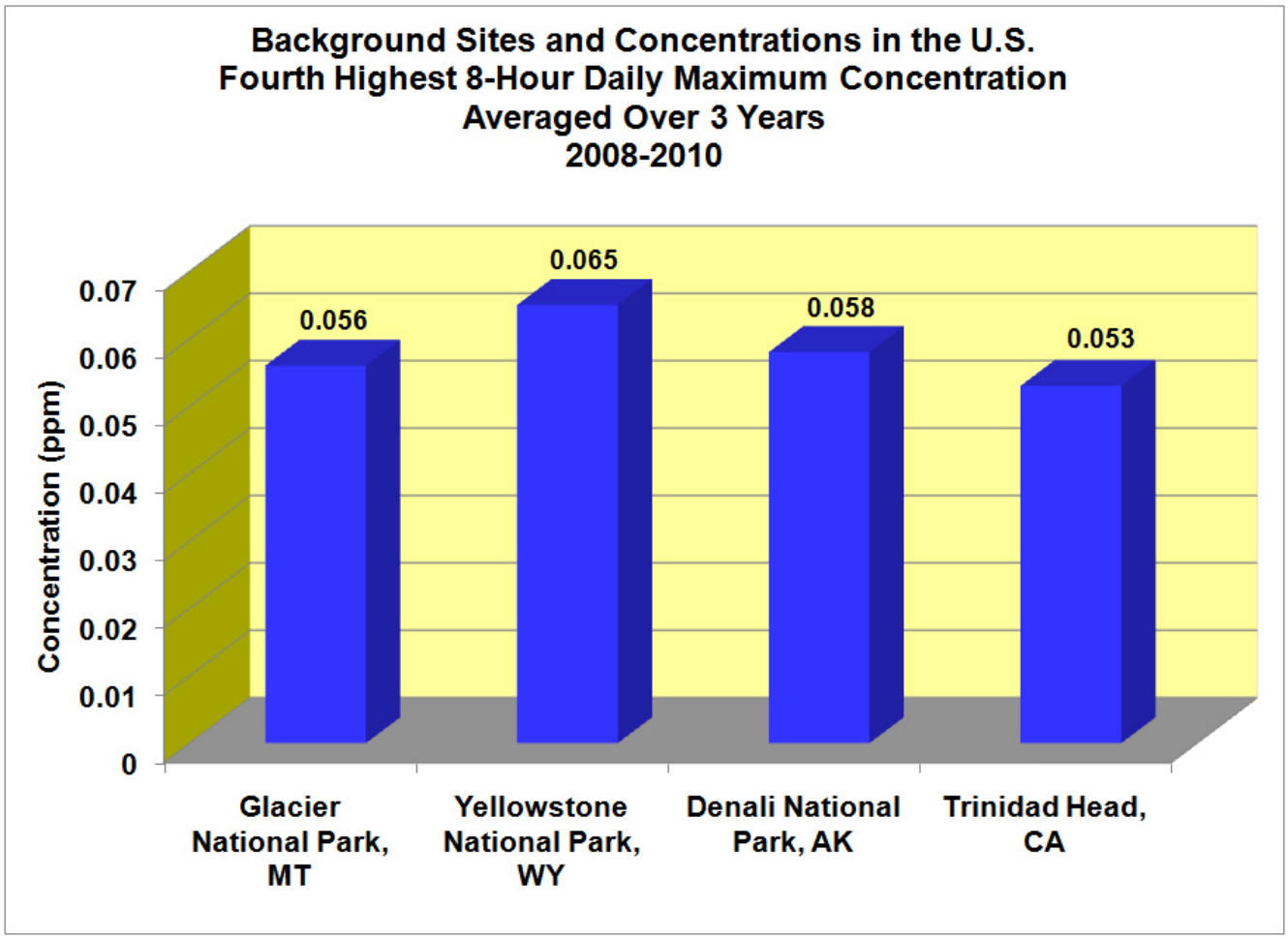

Fig. 13. Four O_3 monitoring sites influenced by PRB long-range transport with their fourth highest 8-h daily maximum concentrations averaged over 3 years for 2008 to 2010. The concentrations are in units of ppm.

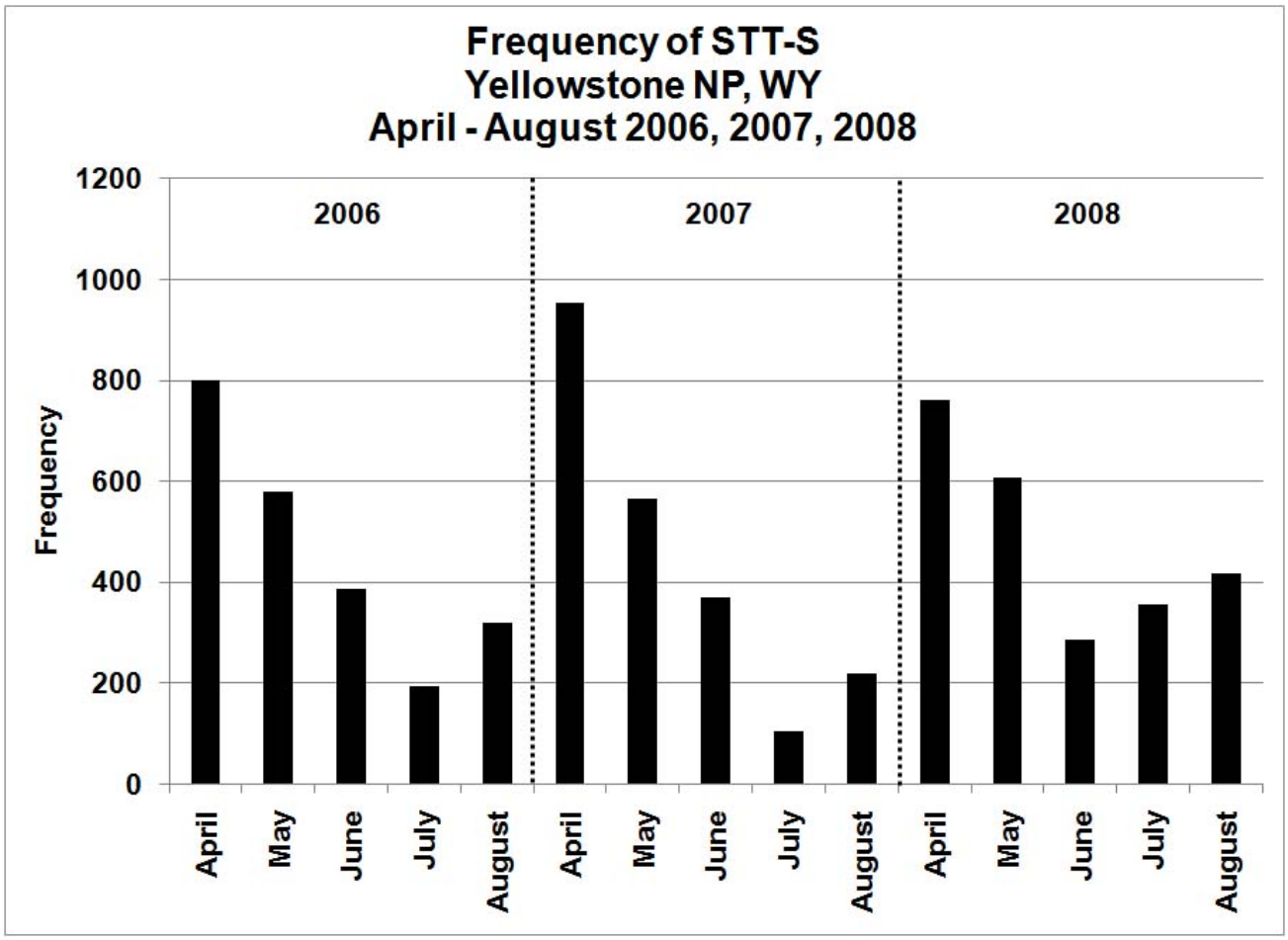

Fig. 14. Frequency of stratosphere-to-troposphere transport to surface (STT-S) at Yellowstone National Park in Wyoming for April – August 2006, 2007, and 2008. Source: Manuscript submitted.

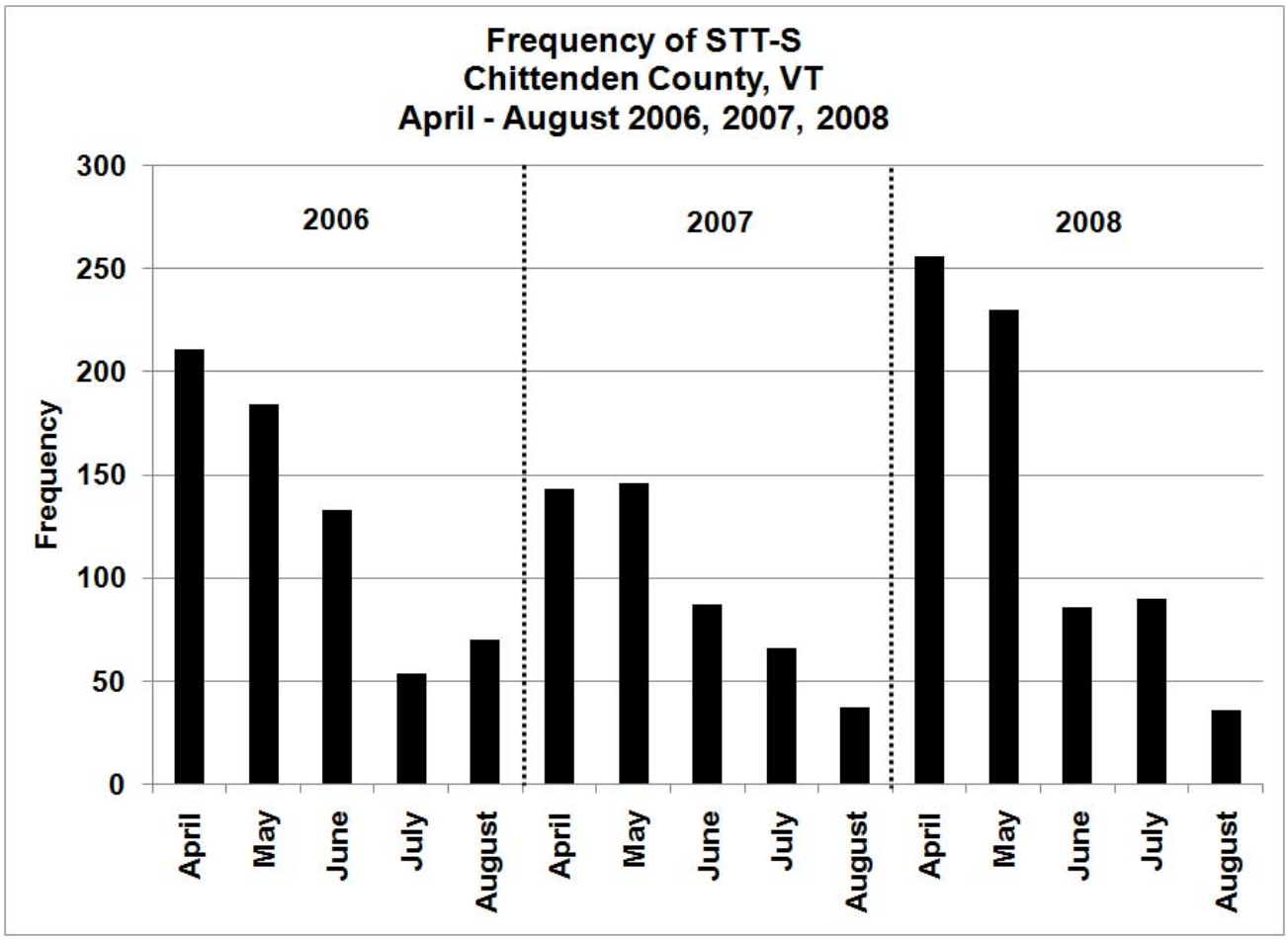

Fig. 15. Frequency of stratosphere-to-troposphere transport to surface (STT-S) at Chittenden County, Vermont for April – August 2006, 2007, and 2008. Source: Manuscript submitted.

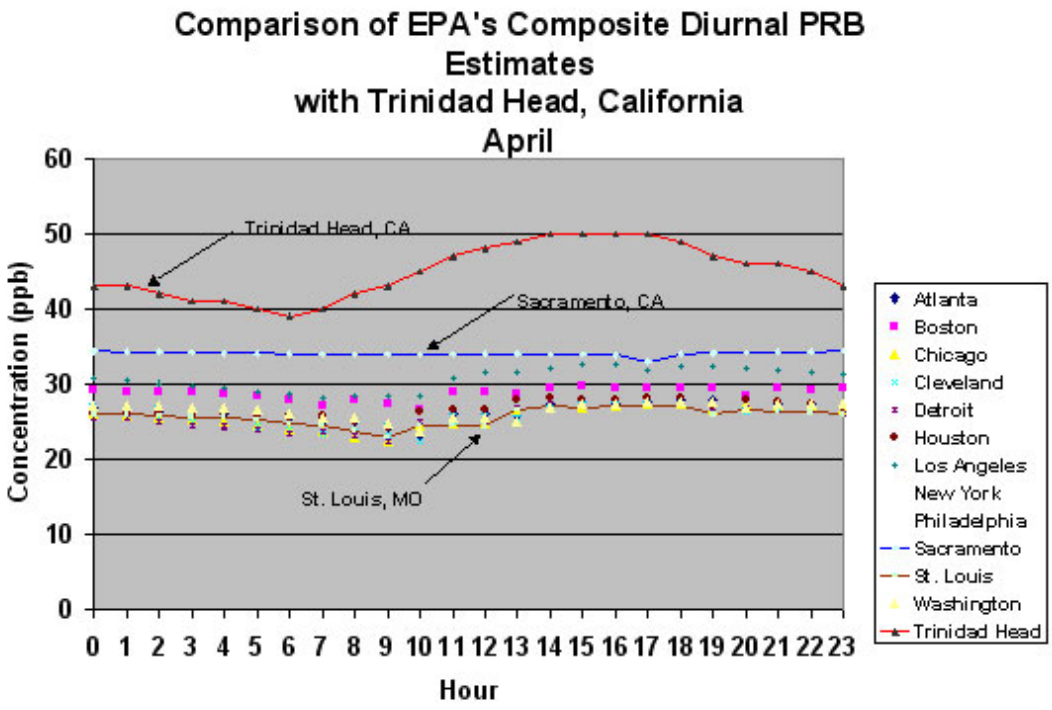

Fig. 16. Comparison of EPA's composite diurnal PRB O_3 estimates with Trinidad Head, CA for April.

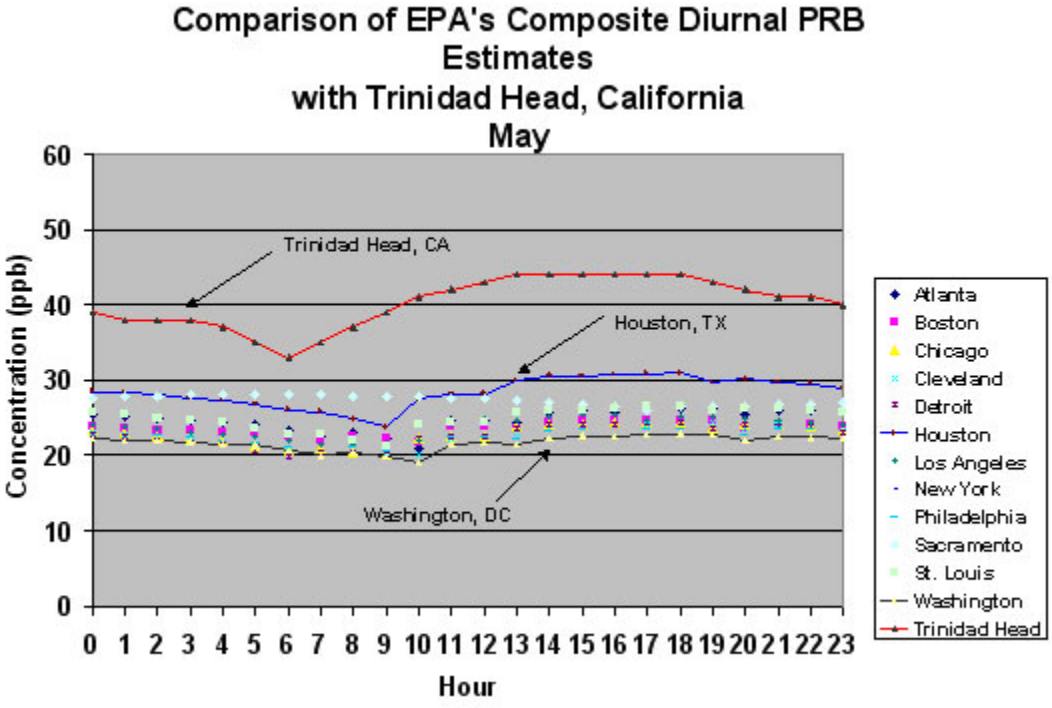

Fig. 17. Comparison of EPA's composite diurnal PRB O_3 estimates with Trinidad Head, CA for May.

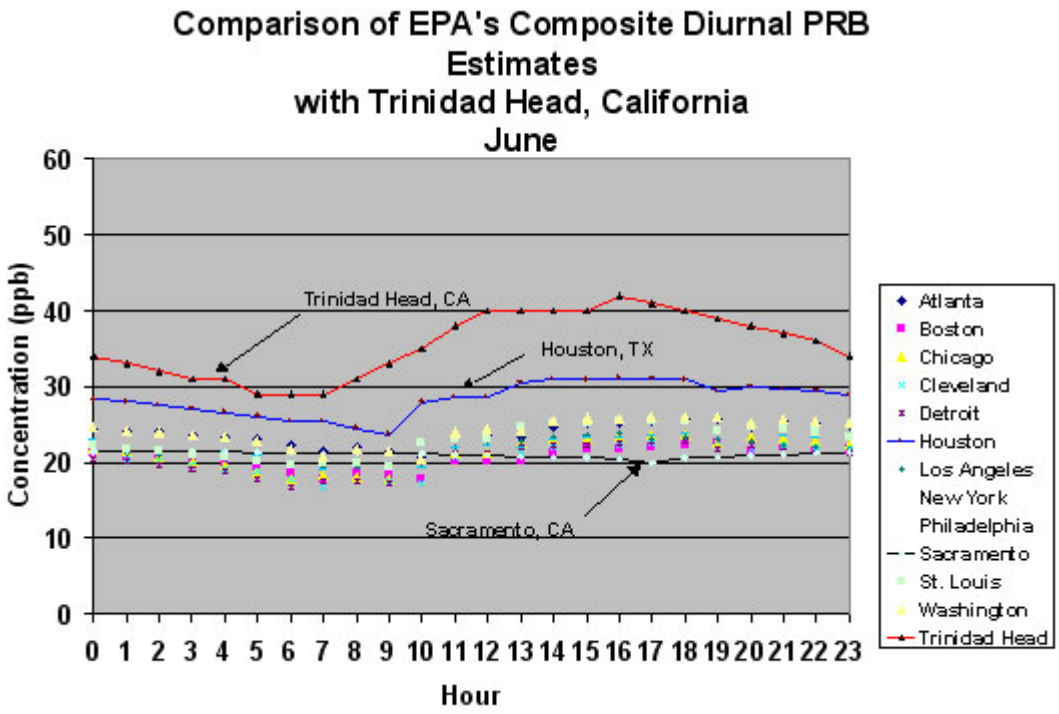

Fig. 18. Comparison of EPA's composite diurnal PRB O_3 estimates with Trinidad Head, CA for June.

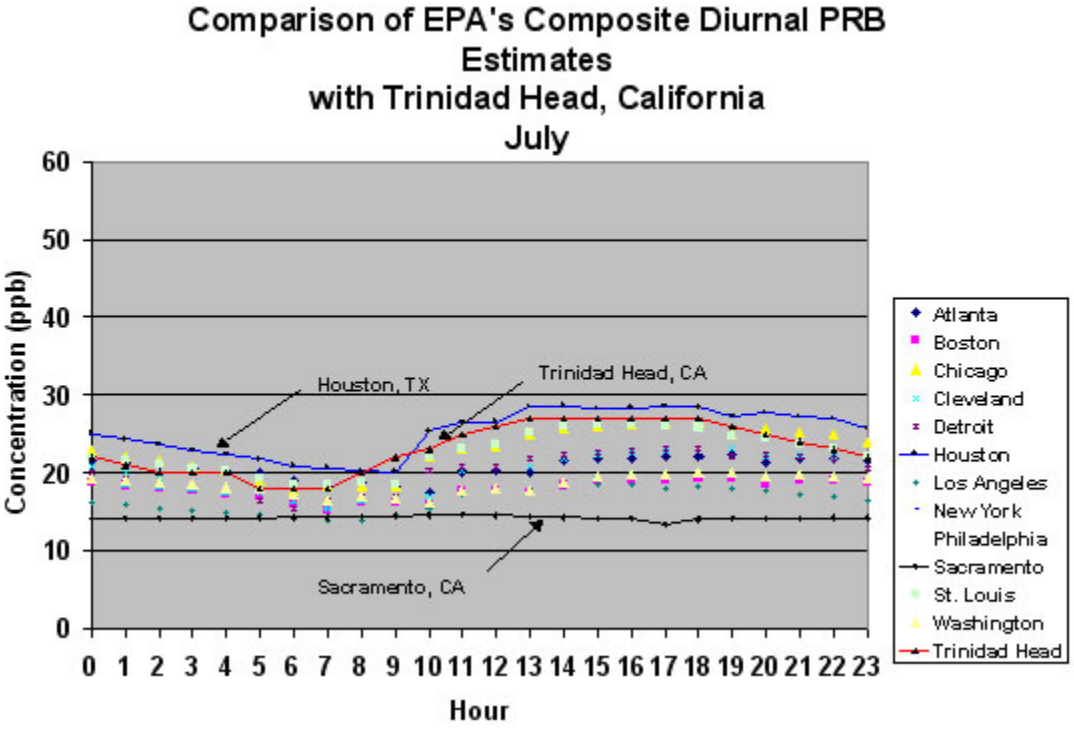

Fig. 19. Comparison of EPA's composite diurnal PRB O_3 estimates with Trinidad Head, CA for July.

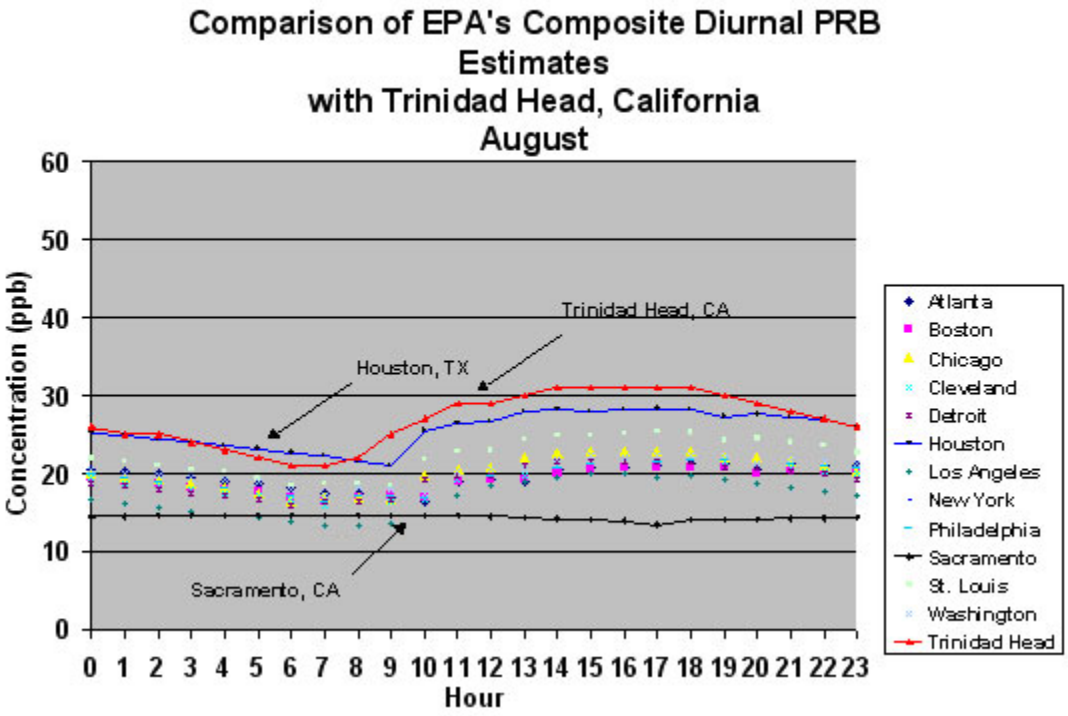

Fig. 20. Comparison of EPA's composite diurnal PRB O_3 estimates with Trinidad Head, CA for August.

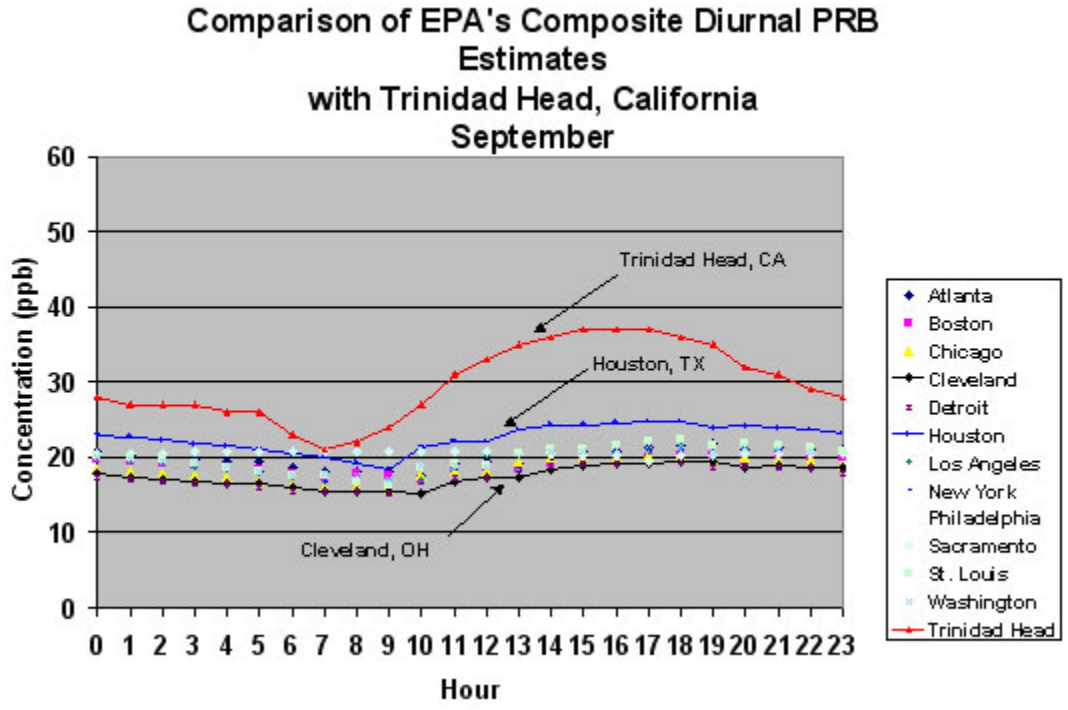

Fig. 21. Comparison of EPA's composite diurnal PRB O_3 estimates with Trinidad Head, CA for September.