Background Ozone Concentrations and the Stratosphere

Comments Relating to the Draft CASAC Panel Reports on ISA, REA, and PA

For Consideration by CASAC

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It has been suggested (pages 10 and 11 of the draft CASAC Panel Report on the ISA) that the importance of the stratosphere be de-emphasized in Figs. 2-1 and 3-1 of the ISA. Furthermore, on pages 11 and 12 of the draft CASAC Panel Report on the ISA, it is suggested that O_3 coming down to the troposphere in stratospheric intrusions is not necessarily natural and could have originated from production in the troposphere and that the high O_3 concentrations observed at elevated monitoring sites reflect the general increase of background O_3 concentrations with altitude in the troposphere due to increasing O_3 lifetime. On page 9 of the draft CASAC Panel Report on the PA, it is also suggested that, other than for the case of exceptional events, the observed increase of O_3 is not associated with the stratosphere. I suggest that CASAC consider modifying the draft wording in its Panel Reports. A more balanced perspective concerning the importance of the stratospheric contribution in respect to background O_3 concentrations is needed in the draft CASAC Panel Reports. I explain below the reasons for my recommendations.

There is considerable evidence published in the literature that the stratosphere is affecting surface O_3 concentrations during the springtime as well as other times of the year. Recent publications include: Lin et al. (2012), Lefohn et al. (2012), Langford et al. (2011), Ambrose et al. (2011), Lefohn et al. (2011), Cooper et al. (2011), and Langford et al. (2009). A separate appendix (Appendix A) is attached that provides the summary abstracts associated with current published papers supporting the importance of stratospheric intrusions and background O_3 influencing hourly average O_3 concentrations $\geq$ 50 ppb.

Besides peer-reviewed research articles discussing the importance of the stratosphere, State and Federal Officials have focused on the importance of the stratosphere affecting surface O_3 concentrations. Recently, recognizing the importance of the stratosphere to affect attainment of the O_3 NAAQS, state air quality agency science and regulatory staff, as well as Federal scientists, met in Reno Nevada to discuss the scientific evidence available for understanding natural background concentrations and the role that background O_3 concentrations play in attainment. Several of the research scientists or representatives from their research group cited in the above paragraph, as well as members of the US EPA, attended the conference. The conference, hosted by the University of Nevada, Reno, and the Western Air Resources Council (WESTAR) was held on October 10-12. The overhead presentations given at the conference can be viewed at

http://www.westar.org/12%20Tech%20Conf/Western%20Ozone%20Transport%20Conference%20presentations.htm

A member of EPA Region 8 indicated that there was compelling evidence that stratospheric O_3 intrusion affected surface O_3 concentrations. The EPA scientist pointed out that the States of Wyoming and Colorado were leaders in documenting the importance of stratospheric O_3 intrusion in enhancing O_3 concentrations. The EPA Region 8 scientist indicated that the stratospheric intrusions that would normally not be considered contributing to exceptional events can contribute to enhanced O_3 concentrations. The EPA is actively participating in the Ozone Stratospheric Intrusion Workgroup whose participants include, besides the EPA, state agencies, NOAA, NCAR, and NASA researchers.

Other participants at the workshop described state agency results that illustrated the importance of stratospheric intrusions affecting surface O_3 concentrations. In addition, several NOAA and other scientists presented empirical and modeling results supporting the importance of the stratosphere in enhancing surface O_3 concentrations.

As I indicated in testimony to CASAC in September, officials in Alberta, Canada note that high springtime O_3 levels are attributable to natural phenomena, such as stratospheric O_3. Extensive research investigations in Alberta have identified the importance of stratospheric intrusion in affecting surface O_3 concentrations at both high- and low-elevation sites. The Province of Alberta believes that the 65 ppb 8-hour average Canada Wide Standard is very sensitive to interference from stratospheric intrusion events. As a result, the Province of Alberta uses an alternative exposure metric, which it believes is less sensitive to interference from stratospheric intrusion, for assessing human health effects.

It is my recommendation, based on my research as well as the peer-reviewed articles published by my colleagues, that a better balanced perspective that reflects the importance of the stratosphere in affecting background O_3 concentrations be presented in the CASAC Panel Reports. The current suggested draft wording de-emphasizes the importance of the stratosphere in affecting surface O_3 concentrations and does not reflect the current state of scientific knowledge on the subject.

It has been suggested (page 2 of the draft CASAC Panel Report on the HREA) that HDDM has the advantage of providing a protocol for the minimum emission reductions needed to achieve just meeting the O_3 standard, and it does not require estimates of "background" O_3. Furthermore, on page 10 of the draft CASAC Panel Report of the HREA, it is stated that "Additionally, background ozone is already incorporated in estimates of <u>change</u> in risk from existing levels to levels that meet the current standard, and is planned to be incorporated in estimates in the next draft involving change in risk for several alternative levels of the standard." Although it might appear to some that it is not necessary to be concerned about estimates of background when the HDDM is applied and the <u>change</u> in risk is estimated, ignoring the uncertainties in O_3 background will lead at times to large uncertainties in the predicted distribution of hourly average concentrations that will be used in the risk assessments. If the HDDM underestimates the importance of natural processes that contribute to background, such as stratospheric-tropospheric exchange, the hourly average concentration distributions resulting from the application of HDDM will result in overly optimistic estimates of health benefits (i.e., the <u>change</u> from existing levels to levels that meet current or alternative standards). Uncertainties in the

distribution of the hourly average concentrations generated by HDDM will be transferred into the estimates of risk. These uncertainties will show up in the change in risk calculations. The issue of properly estimating background O_3 concentrations does not disappear when change in risk is selected as the metric of interest.

References

Ambrose, J.L., Reidmiller, D.R., Jaffe, D.A., 2011. Causes of high O3 in the lower free troposphere over the Pacific Northwest as observed at the Mt. Bachelor Observatory. Atmos. Environ. 45, 5302-5315.

Cooper, O.R., Oltmans, S.J., Johnson, B.J., Brioude, J., Angevine, W., Trainer, M., Parrish, D.D., Ryerson, T.R., Pollack, I., Cullis, P.D., Ives, M.A., Tarasick, D.W., Al-Saadi, J., Stajner, I., 2011. Measurement of western U.S. baseline ozone from the surface to the tropopause and assessment of downwind impact regions. J. Geophys. Res. 11, D00V03, doi:10.1029/2011JD016095 doi:10.1029/2011JD016095.

Langford, A.O., Aikin, K.C., Eubank, C.S., Williams, E.J., 2009. Stratospheric contribution to high surface ozone in Colorado during springtime. Geophys. Res. Lett. 36, L12801, doi:10.1029/2009GL038367.

Langford, A.O., Brioude, J., Cooper, O.R., Senff, C.J., Alvarez II, R.J., Hardesty, R.M., Johnson, B.J., Oltmans, S.J., 2011. Stratospheric influence on surface ozone in the Los Angeles area during late spring and early summer of 2010. J. Geophys. Res. 117, D00V06, doi:10.1029/2011JD016766.

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

Lefohn, A.S., Wernli, H., Shadwick, D., Oltmans, S.J., Shapiro, M., 2012. Quantifying the Importance of Stratospheric-Tropospheric Transport on Surface Ozone Concentrations at High- and Low-Elevation Monitoring Sites in the United States. Atmos. Environ. doi: 10.1016/j.atmosenv.2012.09.004. 62, 646-656.

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

Appendix A

Abstracts of Relevant Papers Detailing the Importance of Stratospheric Ozone and Background Ozone Affecting Surface Ozone Concentrations

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

The published literature debates the extent to which naturally occurring stratospheric ozone intrusions reach the surface and contribute to exceedances of the U.S. National Ambient Air Quality Standard (NAAQS) for ground-level ozone (75 ppbv implemented in 2008). Analysis of ozonesondes, lidar, and surface measurements over the western U.S. from April to June 2010 show that a global high-resolution (~50x50 km2) chemistry-climate model (GFDL AM3) captures the observed layered features and sharp ozone gradients of deep stratospheric intrusions, representing a major improvement over previous chemical transport models. Thirteen intrusions enhanced total daily maximum 8-hour average (MDA8) ozone to ~70-86 ppbv at surface sites. With a stratospheric ozone tracer defined relative to a dynamically varying tropopause, we find that stratospheric intrusions can episodically increase surface MDA8 ozone by 20-40 ppbv (all model estimates are bias corrected), including on days when observed ozone exceeds the NAAQS threshold. These stratospheric intrusions elevated background ozone concentrations (estimated by turning off North American anthropogenic emissions in the model) to MDA8 values of 60-75 ppbv. At high-elevation western U.S. sites, the 25th-75th percentile of the stratospheric contribution is 15-25 ppbv when observed MDA8 ozone is 60-70 ppbv, and increases to ~17-40 ppbv for the 70-85 ppbv range. These estimates, up to 2-3 times greater than previously reported, indicate a major role for stratospheric intrusions in contributing to springtime high-O3 events over the high-altitude western U.S., posing a challenge for staying below the ozone NAAQS threshold, particularly if a value in the 60-70 ppbv range were to be adopted.

Lefohn, A.S., Wernli, H., Shadwick, D., Oltmans, S.J., Shapiro, M., 2012. Quantifying the Importance of Stratospheric-Tropospheric Transport on Surface Ozone Concentrations at High- and Low-Elevation Monitoring Sites in the United States. Atmospheric Environment. doi: 10.1016/j.atmosenv.2012.09.004. 62, 646-656.

In this study, we quantify the frequency of stratosphere-troposphere exchange (STE) events that result in ozone (O_3) concentration enhancements (i.e., hourly average concentrations ≥ 50 ppb) observed at 39 high- and low-elevation monitoring sites in the US during the years 2007-2009. We employ a refined forward trajectory-based approach to address the relationship between stratospheric intrusions and enhancements in hourly average O_3 concentrations. The model is applied to high-resolution European Center for Medium-Range Weather Forecasting (ECMWF) analyses to identify specific days when the potential for stratosphere-to-troposphere transport

(STT) exists to affect surface O_3 levels. Our results indicate that STT down to the surface (STT-S) frequently contributes to enhanced surface O_3 hourly averaged concentrations at sites across the US, with substantial year-to-year variability. The O_3 concentrations associated with the STT-S events appear to be large enough to enhance the measured O_3 concentrations during specific months of the year. Months with a statistically significant coincidence between enhanced O_3 concentrations and STT-S occur 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 exhibit a preference for coincidences during the springtime and in some cases, the summer, fall, and late winter. Besides the high-elevation monitoring sites, low-elevation monitoring sites across the entire US experience enhanced O_3 concentrations coincident with STT-S events.

Langford, A.O., Brioude, J., Cooper, O.R., Senff, C.J., Alvarez II, R.J., Hardesty, R.M., Johnson, B.J., Oltmans, S.J., 2011. Stratospheric influence on surface ozone in the Los Angeles area during late spring and early summer of 2010. J. Geophys. Res. 117, D00V06, doi:10.1029/2011JD016766.

The influence of stratosphere-to-troposphere transport (STT) on surface ozone (O_3) concentrations in the greater Los Angeles area during the CalNex and IONS-2010 measurement campaigns has been investigated. Principal component analysis (PCA) of surface O_3 measurements from 41 sampling stations indicates that ~13% of the variance in the maximum daily 8-h average (MDA8) O_3 between May 10 and June 19, 2010 was associated with changes of 2-3 day duration linked to the passage of upper level troughs. Ozonesondes launched from Joshua Tree National Park and airborne lidar measurements show that these changes coincided with the appearance of stratospheric intrusions in the lower troposphere above southern California. The Lagrangian particle dispersion model FLEXPART reproduces most of these intrusions, and supports the conclusion from the PCA that significant transport of stratospheric air to the surface occurred on May 28-30. This intrusion led to a peak 1-h O_3 concentration of 88 ppbv at Joshua Tree National Monument near the ozonesonde launch site on May 28, and widespread entrainment of stratospheric air into the boundary layer increased the local background O_3 over the entire area to ~55 ppbv on May 29-30. This background was 10-15 ppbv higher than the baseline O_3 in air transported ashore from the Pacific Ocean, and when combined with locally produced O_3 led to several exceedances of the current National Ambient Air Quality Standard (NAAQS) on the following day.

Ambrose, J.L., Reidmiller, D.R., Jaffe, D.A., 2011. Causes of high O_3 in the lower free troposphere over the Pacific Northwest as observed at the Mt. Bachelor Observatory. Atmos. Environ. 45, 5302-5315.

We measured O_3, CO, Hg, sub-micron particle scattering of mid-visible light (ssp), and water vapor (WV) at the Mt. Bachelor Observatory (MBO) in central Oregon, USA since 2004. The data were used to identify sources of large ozone enhancements in the lower free troposphere (FT). A total of 25 high-ozone events, defined as time periods when the 8-h averaged O_3 mixing ratio was >70.0 ppbv, were recorded. The events occurred between early March and late September. For 18 of the 25 event days we were able to identify sources based on (1) relative

enhancements of O_3, CO, Hg and WV compared with the corresponding monthly distributions and (2) supporting data from the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model, the Navy Aerosol Analysis and Prediction System (NAAPS) global aerosol model, and Moderate Resolution Imaging Spectroradiometer (MODIS) satellite-derived active fire counts from the Fire Information for Resource Management System (FIRMS). Our analysis suggests that enhanced O_3 levels during all the identified events were transported to MBO in the FT and were driven mostly by subsidence of O_3-rich air masses from the upper troposphere/lower stratosphere (UT/LS), an long range transport (ALRT) and mixed ALRT+UT/LS influences. The UT/LS events were depleted in CO and total atmospheric mercury (TAM) compared to monthly median values. Levels of O_3 and CO tended to be anti-correlated during UT/LS events, consistent with transport from clean regions in the UT. Conversely, the ALRT events were characterized by concomitant enhancements in CO and TAM with mean values during each event that were >70th percentile values for the months during which the events occurred. Unlike for UT/LS events, levels of O_3 and CO during ALRT events tended to be correlated, consistent with photochemical O_3 production in polluted air masses transported across the Pacific. The mixed ALRT + UT/LS events exhibited characteristics that were intermediate between those of the ALRT and UT/LS events. For the classifiable cases the fractional distribution of each event type was: UT/LS, ~44% (n = 8); ALRT + UT/LS, ~33% (n = 6); ALRT, ~22% (n = 4). In terms of the total number of classifiable high-ozone event hours UT/LS, ALRT + UT/LS and ALRT events accounted for ~52% (t = 85), ~36% (t = 59) and ~13% (t = 21), respectively. The results indicate that downward mixing of O_3-rich air masses from the UT/LS together with trans-Pacific transport of urban/industrial and biomass burning emissions from the Asian continent are the most important mechanisms for delivering high O_3 levels (i.e., 8-h averages >70.0 ppbv) to the lower FT in the Pacific Northwest.

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

Stratospheric-tropospheric exchange (STE) processes contribute at both high and low-elevation monitoring sites to background ozone (O_3) concentrations. This study addresses the importance of stratospheric intrusions contributing to enhanced hourly average surface O_3 concentrations (i.e., ≥50 ppb) at 12 O_3 monitoring stations in the western and northern tier of the US for 2006, 2007, and 2008. The Lagrangian Analysis Tool (LAGRANTO) trajectory model identified specific days when stratosphere-to-troposphere transport was optimal to elevate surface O_3 levels. The coincidences between the number of days with a daily maximum hourly average O_3 concentration ≥ 50 ppb and stratosphere-to-troposphere transport to surface (STT-S> 0) were quantified. The high-elevation site at Yellowstone National Park (NP) in Wyoming exhibited the most coincidences (i.e., more than 19 days a month) during the spring and summer for hourly average O_3 concentrations ≥ 50 ppb with STT-S> 0 of the 12 monitoring sites. At this site, the daily 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. At many of the lower elevation sites, there was a preference for O_3 enhancements to be coincident with STT-S> 0 during the springtime, although summertime occurrences were sometimes

observed. When statistically significant coincidences occurred, the daily maximum hourly average concentrations were mostly in the 50-65 ppb range and the daily maximum 8-h average concentrations were usually in the 50-62 ppb range. For many cases, the coincidences between the enhancements and the STT-S events occurred over a continuous multiday period. Supplementary observations, such as (1) the greater frequency of O_3 concentration enhancements occurring during the springtime versus other times of the year, (2) the elevation dependency of the frequency of enhancements, (3) the year-to-year variability, (4) the timing of the hour-by-hour occurrences of the O_3 concentration enhancements within and across monitoring sites, and (5) the detailed analyses of O_3 enhancement events at specific sites, provide additional support for our modeling and statistical results. Our analysis provides an important step in better understanding the variability of natural background O_3 concentrations. The study has provided insight into stratospheric intrusions, with emphasis on the combined role of quasi-isentropic large-scale advection and mesoscale boundary layer turbulence for stratospheric air influencing enhanced surface O_3.

Langford, A.O., Aikin, K.C., Eubank, C.S., Williams, E.J., 2009. Stratospheric contribution to high surface ozone in Colorado during springtime. Geophys. Res. Lett. 36, L12801, doi:10.1029/2009GL038367.

The contribution of stratosphere-to-troposphere transport to high surface ozone (O_3) along the Colorado Front Range during spring of 1999 is examined using lidar and surface measurements. A deep tropopause fold brought ~215 ppbv of O_3 to within 1 km of the highest peaks in the Rocky Mountains on 6 May 1999. One-minute average O_3 mixing ratios exceeding 100 ppbv were subsequently measured at a surface site in Boulder, and daily maximum 8-hour O_3 concentrations greater or equal to the 2008 NAAQS O3 standard of 0.075 ppmv were recorded at 3 of 9 Front Range monitoring stations. Other springtime peaks in surface O_3 are also shown to coincide with passage of upper level troughs and dry stable layers aloft. These results show that the stratospheric contribution to surface ozone is significant, and can lead to exceedance of the 2008 NAAQS O_3 standards in a major U.S. metropolitan area.

Oltmans, S.J., Lefohn, A.S., Harris, J.M., Shadwick, D., 2008. Background ozone levels of air entering the west coast of the U.S. and assessment of longer-term changes. Atmos. Environ. 42, 6020-6038.

An analysis of surface ozone measurements at a west coast site in northern California (Trinidad Head) demonstrates that this location is well situated to sample air entering the west coast of the U.S. from the Pacific Ocean. During the seasonal maximum in the spring, this location regularly observes hourly average ozone mixing ratios ≥50 ppbv in air that is uninfluenced by the North American continent. Mean daytime values in the spring exceed 40 ppbv. A location in southern California (Channel Islands National Park) demonstrates many of the characteristics during the spring as Trinidad Head in terms of air flow patterns and ozone amounts suggesting that background levels of ozone entering southern California from the Pacific Ocean are similar to those in northern California. Two inland locations (Yreka and Lassen Volcanic National Park) in northern California with surface ozone data records of 20 years or more are more difficult to

interpret because of possible influences of local or regional changes. They show differing results for the long-term trend during the spring. The 10-year ozone vertical profile measurements obtained with weekly ozonesondes at Trinidad Head show no significant longer-term change in tropospheric ozone.

Additional published sources describing the importance of stratospheric O_3 affecting surface O_3 concentrations at both high- and low-elevation monitoring sites during the springtime, as well as other times during the year:

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

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

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

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

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

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

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