

Attributing Sources of Background Ozone

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

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Discussion of the role, levels, and relevance of background tropospheric ozone has been considered in the third draft of the Integrated Science Assessment for Ozone and Related Photochemical Oxidants. The determination of background ozone levels from photochemical models and the fidelity of these modeled ozone levels in comparison with observed ozone amounts under conditions likely to be representative of “background” conditions were presented in public comments to CASAC at its meeting in September 2012. Recently published work by Lin et al. [2012] dramatically reinforces the important contribution of North American background ozone (NAB), including a very significant stratospheric component, on 8-hour maximum daily average ozone (MDA8) at or near current air quality standards. In particular during the spring and early summer, background ozone over the western U.S. is routinely elevated by input from the stratosphere. These findings emphasize the need to provide a balanced view on the contributions to background ozone. In particular several of the responses in the draft of the *Consensus Responses to Charge Question of the Integrated Science Assessment and Policy Assessment* minimize the role of transport of O₃ from the stratosphere in enhancing background ozone when in fact new results suggest that this source plays a larger role than previously thought in background O₃ levels.

The work of Lin et al. [2012] (published paper included as a separate attachment) is a major advance within the modeling framework in the ability to quantify the contributions to background ozone. Unlike prior GEOS-Chem work, the AM3 model explicitly simulates O₃ variability in the lower stratosphere and its dynamic coupling with the troposphere, as opposed to using a parameterized (SYNOZ or LINOZ) formulation or a climatological stratosphere (e.g., as in Fiore et al. [2003] and Zhang et al [2011]). In the *Consensus Responses to Charge Questions on EPA’s Integrated Science Assessment for Ozone and Related Photochemical Oxidants (Third External Review Draft – June 2012)* for Chapter 3 it states:

“1. Figure 3-1 is too complicated for the non-specialist, a comment made on the Second Draft ISA as well. There is no need to show the stratosphere in such detail and it gives the misleading impression that the stratosphere is a large source of ozone for the troposphere.” As will be detailed below, it is important that the ISA portray a balanced perspective on sources that may contribute to background ozone levels including cases where O₃ concentrations are ≥ 50 ppb. The *Consensus Responses* also state:

“2. It should be pointed out that the ozone coming down to the troposphere in stratospheric intrusions is not necessarily natural, i.e., it could have originated from production in the

troposphere followed by transport to the lowermost stratosphere. Lin et al. [2012] considers any ozone that has crossed the chemical tropopause surface as stratospheric; this is a very different definition from that used by Zhang et al. (2011), in which ozone is considered stratospheric only if it was produced in the stratosphere.” In fact, the use of the dynamic e90 tracer in AM3 represents a major improvement in term of avoiding labeling tropospheric ozone as “stratospheric”. The e90 is a surface emitted tracer whose concentration quickly increases when there is direct influence of upward mixing of tropospheric air near the tropopause. As such, AM3 does not account such O₃ as "stratospheric". The extent to which tropospheric air influences baseline O₃ levels that have their origin in the stratosphere is small, especially under the circumstances of deep stratospheric intrusion events. Thus O₃ diagnosed by the AM3 model as coming from the stratosphere is in fact important in influencing surface O₃ concentrations.

Based on AM3, estimates of stratospheric impacts on springtime surface O₃ over the western U.S. are generally higher on average, and up to 2–3 times greater during the intrusions, than previous model estimates [e.g. Fiore et al., 2003]. This finding is in notable contrast to prior work concluding that stratospheric influence on high surface O₃ events is rare [U.S. EPA, 2007]. While some of the discrepancies may reflect the higher resolution of AM3 and interannual variability associated with the El Nino-Southern Oscillation (ENSO), as well as other factors, the AM3 explicit simulation of O₃ variability in the lower stratosphere and its dynamic coupling with the troposphere as opposed to using a parameterized (SYNOZ) or a climatological stratosphere is likely the major reason for the improved simulation of episodic stratosphere to troposphere transport impacts. Prior work has shown a dependence on the attribution of mean stratospheric impacts to the different stratospheric O₃ tagging methods used. The use of the dynamic e90 tracer in AM3 represents a major improvement in tagging stratospheric O₃. It should be emphasized that AM3 simulations of total O₃ and background O₃ (eliminating NA anthropogenic influence), which reproduces observed O₃ enhancements both in surface air and aloft during the intrusions, are independent of the tagging method. This analysis implies that stratospheric intrusions may pose a challenge for springtime O₃ over the U.S. Mountain West to stay below the O₃ NAAQS with domestic emission controls, particularly if a threshold value in the 60–70 ppb range were to be adopted [U.S. EPA, 2010]. Although O₃ produced from local emissions can dominate O₃ pollution in urban areas, in lower elevation U.S. regions, and during summer, nevertheless, stratosphere to troposphere transport may influence surface O₃ over mid latitude regions prone to deep intrusions including during periods when there is O₃ enhancement from pollution sources. These intrusion events are most frequent in the spring but may occur in other seasons.

The influence of O₃ transported from the stratosphere is not limited to exceptional events but makes a contribution to the levels of background O₃ (see figure below). The *Consensus Responses of the Policy Assessment Review* of Chapter 1 states:

“3. The chapter attributes the increased ozone concentrations at higher elevations to stratospheric intrusions. However, other than the case of exceptional events, the observed increase is a result of higher ozone background with higher altitude due to increasing ozone lifetime (drier air) and lack of contact with the surface. Similarly, it should be explained that ozone in the mid-latitudes background atmosphere peaks in spring and is low in summer, primarily due to atmospheric chemistry rather than stratosphere- troposphere exchange.”

While stratospheric O₃ intrusions that lead to the classification of an event as an “exceptional event” are one manifestation of O₃ transport from the stratosphere to ground level, the contribution from the stratosphere to background O₃ levels at more modest levels is very significant. As can be seen in Fig. 1 below, stratospheric O₃ contributes to background O₃ over a wide range of values and particularly at levels ≥ 50 ppb. This important contribution to background levels is important in assessing risk.

In summary the work of Lin et al. [2012] shows that stratospheric intrusions can increase surface MDA8 ozone by 20-40 ppb above baseline levels and generally make a larger contribution to background levels than transported Asian pollution (see Fig. 1 below). This estimate is less sensitive to the tagging method used than previous model simulations, and AM3 simulations of the total O₃ concentrations and background O₃ (eliminating NA anthropogenic influence) reproduce observed O₃ enhancements both in surface air and aloft during the intrusions. The recent work of Lin et al. [2012] along with numerous other published results provide strong justification for providing a balanced view in the Assessment documents of the various contributions to background O₃ for the purpose of assessing risk. With the goal of the ISA, as well as the REA and PA reflecting the most current scientific knowledge, it is important that several statements in the *Consensus Responses* need to be reevaluated in light of this information.

References

Lin, M., A.M. Fiore, O.R. Cooper, L.W. Horowitz, A.O. Langford, H. Levy II, B.J. Johnson, V. Naik, S.J. Oltmans, and C.J. Senff (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.

Fiore, A., D. J. Jacob, H. Liu, R. M. Yantosca, T. D. Fairlie, and Q. Li (2003), Variability in surface ozone background over the United States: Implications for air quality policy, *J. Geophys. Res.*, 108(D24), 4787, doi:10.1029/2003JD003855.

Zhang, L., et al. (2011), Improved estimate of the policy-relevant background ozone in the United States using the GEOS-Chem global model with $1/2^0 \times 2/3^0$ horizontal resolution over North America, *Atmos. Environ.*, 45(37), 6769–6776, doi:10.1016/j.atmosenv.2011.07.054.

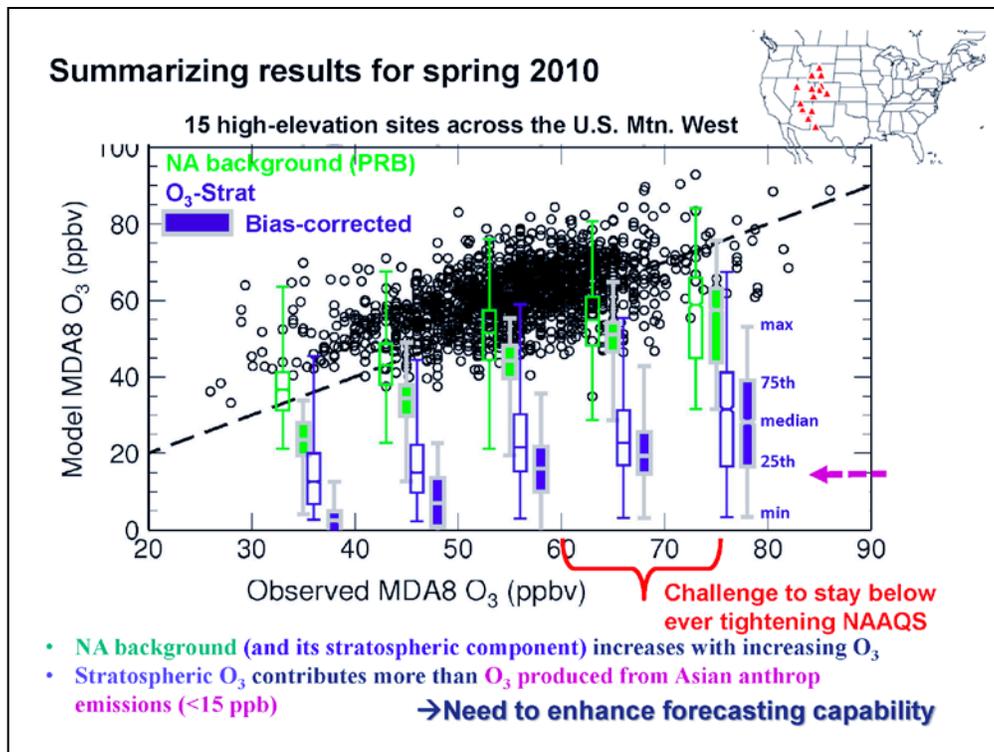


Figure 1: Model versus observed MDA8 surface O₃ for April–June 2010 at 15 high-elevation sites. Also shown is the 1:1 line. The box-and-whisker plots (minimum, 25th, 50th, 75th percentiles, and maximum) give statistics of the NA background (green) and the stratospheric contribution (blue) for every 10-ppb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