Atmos. Chem. Phys. Discuss., 10, C12182–C12187, 2011
www.atmos-chem-phys-discuss.net/10/C12182/2011/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Seasonal and spatial variability of surface ozone over China: contributions from background and domestic pollution" *by* Y. Wang et al.

Anonymous Referee #1

Received and published: 11 January 2011

1 Overview

This paper by Wang et al. investigates the seasonal and spatial trends in surface O_3 over China. In the first step, model simulations of a nested calculation are compared to surface observations, plane flight data and satellite observations. The overall qualitative performance of the model is good, and also quantitative in many aspects, though apparently biased low at times. The next part of this work investigates the sources of O_3 in China using zero-out simulations, attributing total ozone to background vs local anthropogenic vs long-range anthropogenic sources. Lastly, an offline tagged O_x simulation is used to lean more about the region of origin for O_3 that is transported to

China. While several papers in the last two years have addressed long-range transport to China as part of coarser eastern-Asia domains, this is the first study I've seen to look exclusively at China. That being said, some additional comparisons to recent works in the area are warranted. More seriously, I find fault with one of the main conclusions posited by this paper; from my reading of the results it appears that total ozone is more dominated by Chinese sources, both background and anthropogenic, than by pollution from North America and Europe. I wish the authors to clarify these issues, and others outlined below, prior to publication.

2 major comments

1. While many different results are presented in this work, the main focus is on the following: that there is a significant drop in mid-summer surface O_3 concentrations over populated eastern China. They authors explain that this drop comes primarily from a drop in long-range contributions from North America and Europe. I find it odd that this is a focal point of the paper for several reasons.

    (a) First, the "significant drop" doesn't really seem that significant. It's not clear where the number of ~ 15 ppb cited in the abstract comes from. Ozone seasonality in the eastern regions is shown in Figs. 9(c) and (d). Are the authors are getting excited about the slight dip of about ~ 3 ppb in the plot of TO, total ozone, from June to August in panel (c)? This "drop" seems small relative to the vast increase in TO starting in March and running through October which here visibly correlates with the Chinese pollution source, CPO. Or perhaps they are referring to the drop in TO for region NC in panel (d)? But from a quick digitization of the data presented, I calculate that the mean ozone in NC is 45 ppb, and the drop in July is thus only -9 ppb. Why not instead focus again on the larger signal, which would be the 11 ppb spike upwards from the mean in October? The later is clearly driven by a peak in Chinese pollution ozone. So again, I don't clearly see the drop the authors are studying, and instead it seems that the largest features are driven by Chinese pollution ozone.

(b) That being said, let's analyze this drop. The abstract says that it is driven by reduced transport from Europe and North America, but I don't see how that is supported by the results presented. The largest values of any of the source apportionments in SC and NC is from Chinese sources, i.e., the black squares in Fig 10(a) and 10(c) are up to 30-50 ppb during the summer. Meanwhile, the "significant drop" in O_3 in NC from North American and European sources is about 3 ppb each in NC (Fig 10(b)) and 1 ppb in SC (Fig. 10(d)). The total contribution from all exterior sources never comes close to matching the magnitude of the Chinese sources in any of the eastern regions. Rather, by multiplying the regional source attribution in Fig 9 (c) and (d) by the source-type attribution in Fig. 10 (a) and (c), respectively, it seems that the ozone drops in question are driven not by the reason cited in the abstract, but by fluctuations in ozone that are Chinese in origin and background in nature.

While it is true that the results show a decrease in background O_3 during the summer, and that a fraction of this can be linked to North America and European sources, I think this story is just a small ripple atop of a much larger signal, hence it is odd to get so much attention (i.e., the thrust of the abstract). If the conclusions from this work are to have implications for risk assessment and control strategies, then it seems best to focus on the dominant features, which are that the largest signals in TO over China seem linked to fluctuations in Chinese pollution first and background ozone second (frequency decomposition of TO could be used to test this), and that the background ozone signal is driven mostly by local changes, not distant ones.

(c) Finally, the exact numbers here are subjective owing to the definition of the regions used for the analysis. It appears that much of the drop in O_3 in "populated eastern China" is occurring in the part of the SC box that is over the ocean, which will go down in the summer because water vapor increases, not because of long-range transport. If instead, the authors focused on trends in maximum ozone, or ozone levels that exceed an air quality threshold, they would again likely find much greater contribution from local sources, as they themselves mention on page 27864.

2. It would be helpful to include references to and comparisons with several other more recent works investigating long-range transport of O_3 such as those as summarized in the most recent HTAP report, available at www.htap.org, which compiles results from works such as Fiore et al. (2009), Lin et al. 2010, West et al 2009, and Zhang 2009. I suggest the authors consider Table 4.3 of this report and compare their analysis and conclusions in Section 5 to these numbers for the source / receptor relationships. While the HTAP regions are more broad than those considered in the present manuscript, I think the comparisons would still be meaningful. For example, HTAP table also show that relations such as North America to East Asia are at a minimum during the summer. The estimated magnitude is $\sim$1 ppb, for a 20% reduction in North American emissions – how does this compare to the results in Fig. 10?

3. section 3.3, third paragraph: it seems the most drastic model deficiency is the estimates of CO at Linan site, which are significantly lower than the observations, by as much as 400 ppb in January. It is noted later on that general underestimates in CO and O_3 may reflect underestimates in emissions. While I agree that the model generally captures the spatial and seasonal trends, if there is a large bias in the emissions, and that bias is not distributed uniformly among the various sources being analyzed later in the manuscript, then could the authors estimate how much this could impact the findings related to the absolute value of

the contribution of one source relative to another?

4. tagged O_3 simulations: One thing the authors should mention is that this method of attributing sources of O_3 to sources is potentially misleading, as it only tracks the locations where the O_3 was produced, not the location where the emissions which gave rise to this O_3 production are coming from. For example, PAN is transported long distances; upon subsistance back into the boundary layer, it may decompose and lead to ozone formation. This formation would get classified in the offline tagged O_3 analysis as being O_3 from the location where PAN decomposed, rather than the area whose emissions created the PAN in the first place. That being said, I do appreciate that the authors are careful to talk about their results in the correct manner, that is not making assumptions about the origin of the ozone precursors.

5. zero-out simulations for source categorization: How do the authors estimate that the long-term impacts of NO_x on O_3 via CH_4, which are not accounted for in this approach, might impact their analysis?

3 minor comments

- p27855, 13: "different" → "differ"
- section 3.2: it would be helpful for the reader if these site locations could be indicated on one of the regional maps, such as Fig. 7.
- section 3.3, third paragraph: while the qualitative comparisons are illustrative, providing basic quantitative statistics of the bias and correlation of model compared to the observations would strengthen this paper.
- p27861, 10: just to clarify, I suggest "The *nested* model satisfactorily . . . ".

- Figs 5 and 6: It would be nice to see these more clearly. I suggest only including the latitude values on the left most plot, then taking up the remaining white space by making each plot wider.
- page 27873, line 3,4: a few abbreviations crop up here that could be defined (e.g., PRD, YRD, NCP)
- Fig 9: the horizontal axis labels are a bit cramped; I suggest just using the first letter of each month to abbreviate, or write the month names at an angle so that they fit better/

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 27853, 2010.