

We thank the reviewer for careful reading and thoughtful and constructive comments.

Anonymous Referee #1

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 O₃ concentrations over populated eastern China. The 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 ppbv 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 ~ 3ppbv 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.

Response: The 15 ppv decrease mentioned in the abstract is referred to the decrease of total background ozone over east China from its peak in spring to mid-summer (Jul-Aug), labeled as TBO in Fig 9c and 9d, not TO as noted by the reviewer. In Fig 2, surface ozone observations at the surface sites actually show a summertime trough of 15 ppbv or more. Regarding the summertime drop of ozone, we agree with the reviewer that we need to better clarify in the text this phenomenon and the associated changes in different components of surface ozone. This has been done in the revised manuscript as explained below.

The phenomenon is first shown in Fig 2 by observations, in which ozone observations at several surface sites and mountain sites over east China do not show middle summer peaks as expected from the seasonality in solar radiation and natural emissions of ozone precursors (e.g., soil NO_x and biogenic VOCs). For example, there is a 10 ppb decrease of surface ozone at Miyun site (40N lat) from Jun to July, a decrease of 15 ppbv from the May peak to Jun-Jul-Aug at the Lin An site (30N lat), and a decrease of 20 ppbv from spring to Jun-Jul-Aug at the Hok Tsui site (22N lat). In the original discussion paper, we call it ‘the middle-summer drop in surface ozone’ over east China. Compared with ozone seasonality in middle latitude regions of North America and Europe, this is a unique feature of surface ozone over east China and has been well documented in previous studies referenced in the introduction. After reading the two reviewers’

comments, we agreed that it may not be appropriate to call it the ‘middle-summer drop’, as pointed out by reviewer 2 that it looks like a full minimum in all summer months at some sites. In the revised manuscript, the phenomenon is referred to as ‘the summertime trough’ in surface ozone over East China and for simplicity, the magnitude of the summertime trough is defined as the difference between the summertime minimum and the preceding peak level of ozone. For example, the summertime trough at Lin An is of 15 ppbv. We stated explicitly in the text (Sect. 3.2 in particular) that the starting month, duration, and the magnitude of the trough varied by latitude and region.

(b) That being said, let’s analyze the 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 larger value of any of the source apportionments in SC and NC is from Chinese sources, i.e., the black square in Fig 10(a) and 10(c) are up to 30-50 ppb during the summer. Meanwhile, the “significant drop” in O₃ 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 region. Rather by multiplying the regional source attribution in Fig 9(c) and (d) by the source-type attributions in Fig. 10(a) and (c), respectively, it seems that the ozone drop in question are driven not by the reason cited in the abstract, but by fluctuations in zone that are Chinese in origin and background in nature.

While it is true that the results show a decrease in background O₃ 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’s odd to get too much attention (i.e., the thrust of the abstract). If the conclusions from this work to have implications to 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.

Response: As said in the above response, the focus of the paper is to figure out which ozone components leads to the unique feature of the summertime trough in surface ozone over East China (i.e., NC and SC). The source-receptor analysis in Section 4 quantifies total background ozone (TBO), its natural and anthropogenic components (NBO and PBO), and Chinese pollution ozone (CPO). As shown in Fig 9c-d (source-receptor results) and 10a-d (tagged simulation results), although the Chinese sources make a big contribution in total ozone as pointed out by the reviewer, their contribution shows a significant increase from spring to summer, often peaking in late summer, and thus cannot be the driving force to the summertime trough. Only background ozone (TBO) shows a significant decrease in the summer. The simulated summertime trough in TBO has a magnitude of 15 ppbv for both NC (9c) and SC (9d), consistent with the magnitude of the summertime trough observed at the surface sites. That’s why

we stated in the abstract that the summertime trough is driven by the seasonality of background ozone, not by that of the Chinese sources.

As background ozone reflects the composite contribution of all exterior sources, our analysis went one step further, employing the tagged method in Section 5 to identify the source regions of background ozone that leads to the summertime trough. As shown in Fig 10b and 10c, three tagged ozone tracers show a summertime trough, including ROW (Rest of the World), NA, and EU, with a trough magnitude of about 9 ppbv, 2ppbv, and 2 ppbv respectively. Compared with the ROW tracer, the NA and EU component is relatively small and cannot be the driving force to explain the summertime trough of total ozone. So we agree with the reviewer that the sentence in the abstract on the role of North America and Europe is not appropriate. We've made major revision of the abstract in the revised manuscript, as follows: "The summertime trough in surface ozone over eastern China can be explained by the decrease of background ozone from spring to summer (by -15 ppbv regionally averaged over eastern China). Tagged simulations suggest that long-range transport of ozone from northern mid-latitude continents (e.g. Europe and North America) reaches a minimum in the summer, whereas ozone from Southeast Asia exhibits a maximum in the summer over eastern China. This contrast in seasonality provides clear evidence that the seasonal switch in monsoonal wind patterns plays a significant role in determining the seasonality of background ozone over China."

(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 O₃ in "populated eastern China" is occurring in the part of the SC box that is over the ocean, which will go down in 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.

Response: The ocean boxes are excluded from the defined regions and all the results are revised. The analysis of maximum ozone suggested by the reviewer is a good topic for a companion paper, as we pointed out in the conclusion. The purpose of this paper is to understand the regional-scale, mean feature of surface ozone seasonality over East China, the summertime trough feature in particular, not specifically on the contribution of local sources. For the role of the local sources, we've discussed the heterogeneity of CPO (Chinese pollution ozone) in the text: "For all of China, CPO contributes an average of about 20% of TO in the summer. However, the spatial variability in CPO reaches up to 100% compared with only 20% of variability in TO, as CPO accounts for much larger fractions over regions of large local emissions (c.f. Fig 5e and 6e)."

2. It would be helpful to include reference to and comparisons with several other more

recent works investigating long-range transport of O₃ such as those 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 et al 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 be still 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 ~1 ppb, for a 20% reduction in North American emissions – how does this compare to the results in Fig. 10?

Response: The HTAP table suggests the same seasonality of the long-range transport of O₃ from North America (NA) and Europe (EU) to East Asia (EA) as in our study, both at a minimum during the summer and maximum in spring/winter. The HTAP report estimates the full annual-mean contribution (the response to the 20% emission reduction multiplied by five) of NA and EU to EA is 1.1 ppbv and 1.2 ppbv respectively. Our results in Fig 10 give the annual-mean magnitude of the NA and EU tracer to be 1.2 ppv and 1.5 ppbv respectively, consistent with the HTAP report. Despite of the apparent consistency, we caution here that a direct comparison is not so meaningful because the HTAP table gives the mean impact averaged over East Asia as a whole whereas our study looks exclusively at China. We've included a comparison with the HTAP result in Sect. 5 of the revised manuscript.

3. Section 3.3, third paragraph: it seems that most drastic model deficiency is the estimated 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 underestimated in CO and O₃ may reflect underestimated emissions. While I agree that the model generally captures the spatial and seasonal trends, if there is a large bias in the emission, 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?

Response: The accuracy of emission inventories for China has been a great concern for regional and global modeling studies of long-range transport of ozone. CO is a precursor of ozone. CTM's underestimate of CO observations in China is a common problem for chemical transport models. The model evaluation at the Mondy site suggests that our model is probably OK in treating exterior sources. Our model's underestimate of CO inside China indicates that our results might underestimate the contribution of local sources on ozone by underestimating precursor emissions. But given the model's ability in simulating the overall seasonality of both CO and O₃ inside China, we do not expect this model bias to change the main findings of the paper on the driving factors of the summertime trough in surface ozone.

4. Tagged O₃ simulations: One thing the author should mention is that this method of attributing sources of O₃ to sources is potentially misleading, as it only tracks the locations where the O₃ was produced, not the location where the emissions which gave rise to this O₃ production are coming from. For example, PAN is transported long distances: upon subsistence back into the boundary layer, it may decompose and lead to ozone formation. This formation would be classified in the offline tagged O₃ analysis as being O₃ from the location where PAN decomposed, rather than the area whose emission 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 assumption about the origin of the ozone precursors.

Response: The reviewer's point is well taken. The deficiency of tagged ozone simulation has been well recognized in the literature and by the HTAP report in particular. We've already briefly mentioned this issue in the discussion paper. In the revised manuscript, we made it clear to the readers that the tagged O₃ simulation tracks the location of ozone production not that of the precursor emissions: "This approach allows surface ozone over China to be decomposed into components produced from different regions, but does not track the locations of the precursor emissions which give rise to the ozone production."

5. Zero-out simulations for source categorization: How do the authors estimate that the long-range transport impacts of NO_x on O₃ via CH₄, which are not accounted for in this approach, might impact there analysis?

Response: The long-range transport of NO_x will have a relatively large impact on OH radicals and thus affect the abundance and distribution of atmospheric CH₄. These changes lead to long-term reductions in O₃ abundance reflecting the response time of CH₄ (~12 years), which partly offsets the short-term increase in O₃ due to NO_x emission increases. Our model simulations focus on periods of 1-2 years (including model spin-up) and therefore neglect these long-term responses. As a result, our analysis will tend to overestimate background O₃ because the long-term effect of NO_x on O₃ through CH₄ is not accounted for in our approach. However, it has been suggested by the HTAP report [www.htap.org, 2010] that this long-term feedback of NO_x on O₃ through CH₄ is less than 3% for all months and regions and therefore could be negligible for simulations in which anthropogenic emissions of ozone precursors (NO_x, NMVOC and CO) were reduced together. The zero-out simulations in the paper reduce NO_x, NMVOC and CO simultaneously and thus the long-term feedback through CH₄ is negligible. We've added a brief discussion of this issue in Sect 4 in the revised manuscript.

Minor comments

P27855, 13:"different"→"diff"

Response: Corrected.

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.

Response: We added these site locations on Fig 7 in the revised manuscript.

Section 3.3, third paragraph: while the qualitative comparisons are illustrative, providing basic quantitative statistics on the bias and correlations of model compared to the observations would strengthen this paper.

Response: quantitative bias added.

P27861, 10: just to clarify, I suggest “The nested model satisfactorily ...”

Response: Corrected.

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.

Response: Good suggestion. We’ve revised the figures accordingly.

Page 27873, line 3, 4: a few abbreviations crop up here that could be defined (e.g., PRD, YRD, NCP)

Response: We added the definition of the abbreviations in Section 3.2.

Fig 9: the horizontal axis above are a bit more cramped; I suggest just using the first letter of each month to abbreviate, or write the month names at an angle so they fit better.

Response: Corrected.