

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

Anonymous Referee #2

Specific comments

1. The tagged tracer method employed in this study is also applied to East Asia in a recent ACP manuscript, Nagashima et al. 2010 and so the authors should compare quantitatively their results and identify robust conclusions as well as any discrepancies. Their study expands on the Nagashima et al paper by applying emission perturbation simulations and evaluating the model with additional observational datasets. Nagashima, T., T., Ohara, T., Sudo, K., and Akimoto, H.: The relative importance of various source regions on East Asian surface ozone, *Atmos. Chem. Phys.*, 10, 11305-11322, doi:10.5194/acp-10-11305-2010, 2010. In addition to the HTAP referee cited by anonymous referee #1, the authors should also consider the MICS-Asia publications focused on ozone, e.g.: Han, Z., et al. (2008), MICS-Asia II: Model intercomparison and evaluation of ozone and relevant species, *Atmos. Environ.*, 42, 3491 – 3509, doi:10.1016/j.atmosenv.2007.07.031. Holloway, T., et al. (2008), MICS-Asia II: Impact of global emissions on regional air quality in Asia, *Atmos. Environ.*, 42, 3543– 3561, doi:10.1016/j.atmosenv.2007.10.022. The quantitative results discussed in Section 4 and 5 should be compared to these prior studies.

Response: We thank the reviewer for the suggestion. These studies are now referenced in the revision (introduction, sect 4 and 5) and we found general consistency between our tagged analysis and these studies. Please refer to the revised manuscript.

2. Abstract L1-2, I don't see the "mid-summer" drop in any of the observations except Miyun. Figure 2b suggests there is indeed a 15 ppb between Jun and July at Miyun, but at other two sites, it looks like a full minimum in all summer months – Jun-Jul-Aug it is ~15-20 ppb lower than May and Sep at Hok-Tsui and then for Linan, there is a clear May peak, but then the values are not that different from June through Dec, though there is a slight drop (<10 ppb) between May and Jun. Similarly, seasonal cycles do not show a mid-summer drop in 2c. Perhaps more concerning, the model does not even capture the mid-summer drop at Miyun, places the spring peak too early at Hok Tsui, and too late at Linan. It seems that the model is not properly capturing the factors contributing to the timing of the seasonal cycle; the sensitivity simulations could be used to infer which sources are problematic in the model – and this type of analysis would be easier to interpret if the models and observations were shown in the same panel.

Response: The reviewer's point is well taken. Some of the sites indeed show a full minimum in the summer. Also in response to reviewer #1's comments, the phenomenon of interest for ozone seasonality over east China is referred to as 'the summertime trough' in general in the revised manuscript. For simplicity, the magnitude of the summertime trough is defined as the difference between the

summertime minimum and the preceding peak level of ozone. Please refer to our response to reviewer #1 for more details.

In the revised manuscript, we have put the models and observations in the same panel for these sites. The reviewer points out correctly that our model has discrepancies in capturing the exact timing and magnitude of the summertime trough at some sites. The model's discrepancy can be explained by the uncertainties in precursor emissions, the model's relatively coarse resolution, and the fact that the model year is different from observations. Given these factors, we do not expect a perfect match between model and observations. But as far as the general seasonality at each site and the different seasonality between sites are concerned, it is our understanding that the model has a satisfactory performance, which is recognized by reviewer #1. The reason why the model does not capture the mid-summer drop at the Miyun site has been investigated in our previous study (Wang Y et al., ACP, 2008). It is because our model underestimates local cloud optical depths (COD) in July especially during precipitation events. In the revised manuscript, we've added the following discussion of the model's discrepancies: "The discrepancy is probably due to the model's overestimate of ozone in marine boundary layers in summer [Liu et al., 2006] and the model's inability to simulate cloud optical properties associated with the summer monsoon [Wang Y et al., 2008]. The former will lead to a positive bias in the model's estimate of background ozone in summer and the latter will result in a positive bias in the model's estimate of ozone production from local sources."

3. Abstract L6-7, "Anthropogenic background" should be defined, and used consistently throughout the text (Pollution Background Ozone elsewhere). This seems somewhat over-emphasized from Figure 9, which, for example suggests a fairly constant PBO.

Response: We changed "anthropogenic background" to "pollution background ozone" in the abstract for consistency. Our interpretation of PBO in Figure 9 is that it shows a fairly evident summertime trough in all four regions. Nevertheless, we've rephrased this sentence in the abstract without calling it distinct seasonality: "Pollution background (annual mean of 12.6 ppbv) shows a minimum in the summer and maximum in the spring".

4. Abstract L12-13. Was this sentence intending to refer to west China (Figure 9a) where there does appear to be modeled 15 ppb decrease in TBO (though only ppb TO)?

Response: This sentence is referred to east China, that is Fig 9c and 9d.

5. It's not clear where 2001 or 2006 is used for the background ozone analysis. Whichever year it is, consider evaluating that year in Section 3, or include both years in each plot if interannual variability plays a strong role.

Response: We used 2006 for the background ozone analysis in Sect. 4. We

evaluated 2006 nested-grid model results in Section 3.2 (Fig 2) and 3.4 (Fig 4). The comparison with the TRACE-P aircraft campaign conducted in 2001 is with the nested-grid model results for 2001 (Fig 3). We've clarified this issue in the revised manuscript.

6. Section 2: What is the time frame for the simulations? Spin-up? The first part of Section 4.1 would probably fit best here. Is the soil NO_x only seasonally resolved or computed each time?

Response: The simulations are conducted for the full year of 2006 after a 12-month spin-up (Jan-Dec 2005) of the global model. We described the simulation time frame and spin-up in Sect. 2 and moved the first part of Sect 4.1 to Sect 2. The soil NO_x is computed at each model time step except for east China where the soil NO_x emissions are only seasonally resolved following the inversion study of Wang et al. (2007a).

7. Section 3.2: L6-7 p27860: Mt Tai also features a secondary peak in fall. L9-10 it's not clear this statement is accurate for Linan, Miyun, Mt Hua. L18-19 Explain further how this influence the surface ozone simulation. P.27861 L10 perhaps rephrase to "the model is sampled along the flight tracks each minute"?

Response: L6-7 Corrected. L9-10 we mean general features of the seasonality. L18-19 we added the following discussion: "The former will lead to a positive bias in the model's estimate of background ozone in summer and the latter will result in a positive bias in the model's estimate of ozone production from local sources." Also see our response above. L10 corrected.

8. Section 3.4: For these comparisons, is there a selection for land only? I don't understand why the data is separated into east/west China in this way; the seasonal cycles look fairly similar, and it seems more appropriate to separate north/south given the different seasonality at lower/higher latitudes shown in Figure 2. If you compare the E China in Figure 4 with the mountain sites, how similar are they, and can you conclude something about the spatial representativeness of those mountain sites?

Response: Yes, there is a selection for land only. The reason why we separate the data into east/west is because the contribution of Chinese emissions on the tropospheric ozone budget is much larger in the east than the west. There are also distinct differences in topography and landscape between east and west China (c.f. Fig 7). The data in Fig 4 are tropospheric ozone column which cannot be directly compared with surface ozone concentrations at the mountain sites.

9. Section 3.5 L14-16. This statement seems to assume that chemistry and transport are well represented in the model. How was that determined?

Response: CO has a relative long lifetime and can be transported to long distances. As our model has a good agreement with the seasonality of CO observation (Sect 3), we can establish confidence in our model's ability to represent the transport in the troposphere. The seasonality of ozone is

determined by both transport and chemistry. Our model reproduces the general seasonality of ozone at each site and the different seasonality between different sites (Sect 3), lending confidence to our model's ability to simulate the ozone chemistry.

10. Section 4.1. The first paragraph does not fit under "Mean background". The relationships in L21-22 hold by definition; are they expected to be linearly additive or is this interpretation confounded by non-linear chemistry? L27-28. It's not clear why Fiore et al findings are relevant here. In addition to the TF HTAP papers (see referee #1) and others cited above, consider: Fiore et al 2003 and Wang H. Q. et al (2008) cited in the text elsewhere; note in particular the Wang et al study finds higher background when Canadian and Mexican emissions are included, which may be more relevant to the emission perturbations conducted here where only Chinese (not all Asian) sources are set to zero. Also, Auvray, M., and Bey, I.: Long-range transport to Europe: Seasonal variations and implications for the European ozone budget, J. Geophys. Res., 110, D11303, 10.1029/2004jd005503, 2005.

Response: We've moved the first paragraph to Sect. 2. The relationships are not expected to be linearly additive because of the nonlinear ozone chemistry. As background ozone is a model construct and cannot be measured directly (HTAP report), this relationship holds by definition based on the emission perturbation modeling methodology. In the revised manuscript, we've referred to the studies recommended by the reviewer: "The seasonality of TBO is consistent with that of long-range transport to East Asia, Europe and North America reported by previous studies [Fiore et al. 2003; Auvray and Bey, 2005; HTAP, 2010]. As the present study zeros out only Chinese anthropogenic emissions, mean background ozone derived over China is higher than background ozone over East Asia suggested by the HTAP study that zeros out all East Asian anthropogenic emissions. Similarly, Wang et al. [2008] found higher background over the US when Canadian and Mexican emissions are included."

11. Section 4.1 p. 27864 L6 – refer to figures here to illustrate? Also L11-12 should be possible to quantify this with the model simulations so there shouldn't be a need to speculate.

Response: L6 -- We referred to Fig 6b. L11-12, we used model results in Fig 5e and 6e to confirm this sentence.

12. Section 4.2 TO seems highest in spring so why not focus on that? Might be more interesting to show two different seasons rather than annual mean. P.27865 L8. Table 2 only shows a 3 ppb difference in PBO between summer and annual mean. L16-17. Was this attribution done with the tagged tracer simulation or is this an assumption? This and the next text sentences are confusing – so which factor is it? L20-24 Was this point confirmed for the simulations presented here for China?

Response: The reason why we show annual mean figures is for easier comparison with previous studies (including the references suggested by the reviewer) which

reported annual mean results. This study focuses on the summertime trough of surface ozone over east China so that's why we showed JJA results.

L8: The numbers corrected.

L16-17: it's done with the tagged tracer simulation. Both factors (stratospheric influence and longer ozone lifetime) contribute to the higher NBO over West China. We've clarified this issue in the revised manuscript: "There are two topography-related factors responsible for the relatively high NBO and thus TBO over west China. First, it can be attributed to stratospheric influences because of the high altitude. This is supported by the tagged ozone simulation in Section 5. Second, it can be explained by spatial variability in ozone lifetime."

L20-24, Yes, as shown in Fig 8.

13. P. 27867 L8-9 also the longer O₃ life time as shown in Wang et al. 1998c: Wang, Y., Jacob, D. J., and Logan, J. A.: Global simulation of tropospheric O₃-NO_x-hydrocarbon chemistry 3. Origin of tropospheric ozone and effects of nonmethane hydrocarbons, *J. Geophys. Res.*, 103, 10757-10767, 10.1029/98jd00156, 1998. Note that Wang et al is also the appropriate reference for first implementing the tagged Ox tracer method (Section 5 cites Fiore et al).

Response: We added this reference in Sect. 4.3 and Sect. 5.

14. P. 27867 L28. Also the circulation does not carry CPO from the dense emissions regions in E China to the west?

Response: The transport of CPO from E China to WC is shown in Fig. 10g, but this component is smaller than CPO from local sources over WC. We've clarified in the text: "..... as local anthropogenic sources are minimal over WC and the contribution of CPO from the dense emission regions in the east to WC is estimated to be less than 5 ppbv at the surface according to the tagged ozone simulation in Sect. 5."

15. P. 27868 L8-9. This looks more like up to 25 ppb for CPO over NC and 20 ppb over SC? Not clear for the sentence, "The relative important... is more complicated" means? P27868 L14-15, are these also off by 5 ppb, e.g., 20→15 and 30→25? This repeats some of what was above. L22 is this really significant? Jul-Aug-Sep all look similar in TO. See points above related to discussion of this in the abstract.

Response: The numbers corrected. That sentence removed and we added the following sentence: "NC and SC have dense population and higher level of economic development, so we discuss the seasonality of CPO and TBO over these two regions in more detail." We meant the decrease of CPO from Jun to July. The abstract is revised. See our response above.

16. p 27871 L13-16 Is this referring to a similar spatial pattern? The seasonal timing differs. This seasonality could be compared to that found in the HTAP and other studies.

Response: Yes, we meant a similar spatial pattern. We've clarified in the text and

added comparison with the HTAP study.

17. P 27872 L3-4 This is not obvious from Figure 10 for SC or NEC. Further, I'm not convinced this annual mean RPI diagnostic is meaningful (except perhaps for WC). Consider for example the NC region – EU may be most important in all months for NC but NA contributes nearly as much, and based on Fiore et al 2009, the relative importance of a source region to a receptor region can vary substantially across models. The other regions in Fig 10 clearly show that different source regions dominate in different months, so is an annual diagnostic useful?

Response: The reviewer's point is well taken. We've deleted the discussion of RPI in the revised manuscript.

18. P 27872 L6-8 – The PA is hard to see and it is too small compared to other influences, is it meaningful? Further, it seems highest at NEC in spring in contrast to what is stated? L12; spring→summer instead of spring→winter? L19-22, point is not clear.

Response: The PA component is removed from the figure. The text is revised.

19. Section 5 and 6, ROW is the largest contributor in the tagged simulations, are there regions that weren't tagged that might contribute larger amounts than the individual regions included here?

Response: No. In the tagged simulation, we defined almost all the regions which would have a great influence on the ozone distribution in China, including neighboring countries such as Japan, India and South Asia, and long-range transport regions with large emissions such as North America and Europe. ROW is the total of all other world regions not defined.

20. Conclusions p27874 L21-22. I do not see how the statement that EU has the largest influence average over China is correct – it is not larger than ROW (Fig 10), or CPO (most months) or NBO (in any month) in Figure 9?

Response: This statement removed.

Technical comments

Table 1: Define the abbreviations used throughout the text here.

Response: Done.

Table 2: Explain how the decomposition is done, and also the region boundaries use to China.

Response: The decomposition and region boundaries are described in the text and in Fig 5a. We added a footnote to the table.

Figure 2: This would be easier to interpret if the models and observations were in the same panel; consider showing each site separately, and possibly modes for 2001 and 2006. Also shown on one the maps where these sites are located.

Response: Done. Fig 2 revised and the site locations are shown in Fig 7.

Figure 4: Why average TES over 3 years rather than compare directly for 2006?

Response: TES instrument has a footprint of and the global repeat time of 14 days, so the spatial coverage of TES data is poor for one year's observation. An average of three years' TES data significantly improves the statistical representativeness of TES data for general seasonality.

Figure 10: Use a different symbol/color for India so it's not interpreted as JaKr as in the other panels.

Response: Done.

Figure 8: color scale could be capped at 7 or 9.

Response: Done.

p. 27863 L 17 Enhancement → ozone to match CPO label.

Response: Corrected.

p. 27863 L 24, Define what “spatial variability” means – is this the standard deviation of all model grid cells falling within the region? Land only?

Response: We've clarified in the text: “...defined as the standard deviation of all grid cells falling within the region, including only the land areas.”

p. 27866 L22 the point of the final sentence is not clear.

Response: Now we change the sentence to “Northeast China is at higher latitude with humid temperate climate, and is most easily influenced by European pollutions due to its latitude.”

p. 27867 L2, Seem more like the Hok Tsui site in that panel peak is larger than the spring peak.

Response: We agreed, but our statement discusses both peaks which both sites feature.

p. 27869 L9 Why does temperature above freezing matter for ozone production? Isn't is more driven by the solar radiation?

Response: We removed the temperature part in this sentence.

p. 27871 L27. awkward wording.

Response: The sentence revised to “JaKr O₃ exerts the largest influence over NEC in the summer and over NC/SC in the fall, as a result of the change in prevailing wind directions.”

Conclusions section could be more quantitative and define regional abbreviations.

Response: Revised.