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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 #2

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General comments

While the approach is scientifically sound, the text requires major revisions prior to publication. I agree with anonymous referee # 1 that major conclusions – firstly the point that observations show a drop in mid-summer over eastern China, and secondly, the interpretation of this "drop" with the model, and thirdly, the statement in the conclusions that EU has the largest influence on Chinese surface ozone - do not seem substantiated by the analysis. It also seems, on the basis of this and prior publications, that the monsoonal circulation is a major factor in determining the seasonality of _total_ surface ozone over China, not just the background ozone asserted in the final sentence of the

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abstract. The manuscript should better place the contributions here in proper context of recent literature. Throughout the text, the language should be revised to improve clarity.

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., 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 references 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 Sections 4 and 5 should be compared to these prior studies.
- 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 drop between Jun and Jul at Miyun, but at the other two sites, it looks like a full minimum in all summer months June-Jul-Aug it is \sim 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 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.

- 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
- 4. Abstract L12-13. Was this sentence intending to refer to west China (Figure 9a) where there does appear to be a modeled 15 ppb decrease in TBO (though only 10 ppb TO)?
- 5. It's not clear whether 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.
- 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 NOx only seasonally resolved or computed each time step?
- 7. Section 3.2: L6-7 p 27860: 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 influences the surface ozone simulation. P. 27861 L10 perhaps rephrase to "the model is sampled along the flight tracks each minute"?
- 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

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something about the spatial representativeness of those 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?
- 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 the 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.
- 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.
- 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 L 8. 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 sentence are confusing so which factor is it? L20-24 Was this point confirmed for the simulations presented here for China?
- 13. p. 27867 L 8-9 also the longer O3 lifetime 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).

- 14. P27867 L28 Also the circulation does not carry CPO from the dense emissions regions in E China to the west?
- 15. P27868 L8-9. This looks more like up to 25 ppb for CPO over NC and 20 ppb over SC? Not clear what the sentence, "The relative importance...is more complicated" means.
- 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. 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.
- 17. p27872 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?
- 18. p.27872 L6-8 the PA is hard to see and it is so 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.
- 19. Sections 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?
- 20. Conclusions p27874 L21-22. I do not see how the statement that EU has the C12231

largest influence averaged over China is correct – it is not larger than ROW (Fig 10), or CPO (most months) or NBO (in any month) in Fig 9?

Technical comments

- Table 1: Define the abbreviations used throughout the text here.
- Table 2: Explain how the decomposition is done, and also the region boundaries used for China.
- 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 models for 2001 and 2006. Also show on one of the maps where these sites are located.
- Figure 4: Why average TES over 3 years rather than compare directly for 2006?
- Figure 10: Use a different symbol/color for India so it's not interpreted as JaKr as in the other panels.
- Figure 8: color scale could be capped at 7 or 9.
- p. 27863 L 17 enhancement -> Ozone to match CPO label.
- p. 27863 L24. Define what "spatial variability" means is this the standard deviation of all model grid cells falling within the region? Land only?
- p. 27866 L22 the point of final sentence is not clear.
- p. 27867 L2. Seems more like the Hok Tsui site in that the fall peak is larger than the spring peak.
- P 27869 L9 Why does temperature above freezing matter for ozone production? Isn't it more driven by the solar radiation?
- P 27871 L27 awkward wording.

Conclusions section could be more quantitative and define regional abbreviations.

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