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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Constraints on Asian ozone using Aura TES, OMI and Terra MOPITT" *by* Z. Jiang et al.

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We thank the reviewer for the thoughtful and detailed comments. Below we respond to the individual comments. In addition to the revisions discussed below, all figures (Figure 4, 5, 6, 7) and tables (Table 2), associated with sensitivity calculation, are recreated with the updated NOx and CO emission inventories. Its influence on the analysis is small.

General comments

(1) The opening sentence of the abstract sets the stage for a discussion of the rising Asian anthropogenic ozone precursors on tropospheric ozone, yet this is absent from much of the rest of the paper. From Figure 2, the year-to-year increases in ozone

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and CO in June and to some extent July over E China do not seem to be transported into the outflow region. Does this imply that even if Asian emissions are increasing, the impact is regional and not detectable downwind? Some discussion of the findings of this paper in the context of rising emission trends seems warranted. What are the reported changes in Chinese emission from 2006-2010 and to what extent are these incorporated into the model?

Response: In the period of Jun – Aug, East Asia is strongly affected by East Asian monsoon. The pollution was transported more northwardly rather than eastwardly, and consequently, the correlation between Eastern China and China outflow is low. However, according to our test, the effect is still detectable downwind.

Most recent studies believe the annual growth rate of Chinese NOx emission is 5-10%. In the GEOS-Chem model, the Eastern China (107.5-122.5E 20-44N) annual anthropogenic NOx emission is 16.5Tg (2006) and 20.7Tg (2010), with a 5% annual growth rate. These numbers have been added in the discussion.

(2) It's not clear how much data is actually available from TES to construct the regional monthly means examined here. Table 1 should include the sample sizes. Is the model being sampled consistently with TES? Are any revisions to understanding based on earlier ozone-CO work needed based on the findings here?

Response: The model values are sampled at TES measurement time and location. Because TES data is sparse, the uncertainty in the monthly slope/correlation could be large. In a recent work, Voulgarakis et al. (2011) used multi-year seasonal mean value. Similar as Voulgarakis et al. (2011), we also calculated mean value, which is more reliable than the monthly mean.

The major objective of this work is the partition between natural and anthropogenic O3 precursors. However, we need to ensure the chemical transport model can provide a good O3 simulation. The analysis about O3-CO slope/correlation and the top-down CO/NOx estimations are all designed to evaluate the model. The analysis about O3-

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CO slope/correlation itself has no significant difference with previous works.

Voulgarakis, A., Telford, P. J., Aghedo, A. M., Braesicke, P., Faluvegi, G., Abraham, N. L., Bowman, K. W., Pyle, J. A., and Shindell, D. T.: Global multi-year O3–CO correlation patterns from models and TES satellite observations, Atmos. Chem. Phys., 11, 5819–5838, doi:10.5194/acp-11-5819-2011, 2011.

(3) How do the findings for 2006 regarding model biases, and the emission updates needed to match the satellite data, compare with earlier studies using satellite and aircraft data from the NASA INTEX-B field campaign (April-May 2006)? In general, more context could be provided to state clearly the new contributions from this paper beyond prior work focused on this region.

Response: Kopacz et al. (2010) did a CO inversion using the same emission inventory as this work. Using the identical inversion approach as this work, we (Jiang et al. 2014) compared the inversion results and found the seasonal variation of the a posteriori combustion emissions of Jiang et al. (2014) is consistent with the results of Kopacz et al. (2010). A complete discussion about the emission inventories may not be suitable here as it is beyond the scope of this paper.

Kopacz, M., Jacob, D. J., Fisher, J. A., Logan, J. A., Zhang, L., Megretskaia, I. A., Yantosca, R. M., Singh, K., Henze, D. K., Burrows, J. P., Buchwitz, M., Khlystova, I., McMillan, W. W., Gille, J. C., Edwards, D. P., Eldering, A., Thouret, V. and Nedelec, P.: Global estimates of CO sources with high resolution by adjoint inversion of multiple satellite datasets (MOPITT, AIRS, SCIAMACHY, TES), Atmos. Chem. Phys.,10, 855-876, doi:10.5194/acp-10-855-2010, 2010.

Jiang, Z., Jones, D. B. A., Henze, D., Worden, H: Sensitivity of inferred regional CO source estimates to the vertical structure in CO as observed by MOPITT, Atmos. Chem. Phys. Discuss., 14, 22939-22984, doi:10.5194/acpd-14-22939-2014, 2014.

(4) The seasonal variation in the ozone chemical regime discussed here could be com-

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pared with earlier work (e.g., Martin, R. V., A. M. Fiore, and A. Van Donkelaar (2004), Space-based diagnosis of surface ozone sensitivity to anthropogenic emissions, Geophys. Res. Lett., 31, L06120, doi:10.1029/2004GL019416) Has there been a change detected in the seasonality of the ozone chemistry due to changing emissions over the past decade(s)?

Response: Thanks for this interesting paper! Martin et al. (2004) showed that the chemical regime over northeast China near-surface levels is VOC limited in summer and NOx limited in winter. By checking the O3 sensitivity in planetary boundary layer (PBL), this newer version of the GEOS-Chem model (relative to the model used in Martin et al.) shows that the chemical regime over northeast China PBL is NOx limited in summer and VOC limited in winter, which is opposite with Martin et al. (2004). The reason for this difference is an interesting topic. However, discussion about the O3 sensitivity in PBL in detail is beyond the scope of this paper as here we are focused on free-tropospheric ozone.

Specific comments

(1) What year-to-year scaling is applied to the Chinese emissions from the INTEX-B 2006 base year inventory? Could the increases in the red line in June & July over E China be solely due to meteorological variability?

Response: The annual growth rate of Chinese anthropogenic NOx is 5% in the model. From Figure 2, the annual O3 increase in June (E China) is about 3%, which seems larger than the expectation based on NOx increase. However, it is not a good idea to evaluate O3 trend by using a single month. Because TES data is sparse, temporal and spatial representation errors could exist in the monthly mean. The influence of meteorological variability on the O3 variation is an interesting question, which we hope to study in our next step.

(2) The abstract comments on the seasonal variation of ozone; why not also the inter-annual variation? In the final sentence of the abstract, is this conclusion drawn from

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anthropogenic NOx contributions shown in Table 2 for the China outflow region? If so, it doesn't seem to hold for summer.

Response: Changed.

(3) Is the 3-7 ppb noted here for Asian emission increases on western North American free tropospheric ozone referring to the 2003 to 2010 period? If so, what is the increase expected for the 2006-2010 period examined here, and is this consistent with Figure 2, and to what extent does it require chemical production to occur during trans-Pacific transport? Some discussion would need to be added to support the statement in the summary asserting that the emissions from ROA and E China influence background ozone over North America. Given the current analysis, that statement appears speculative and should be removed if evidence is not added to support it.

Response: The previous studies (Zhang et al. 2008 and Brown et al. 2011) focused on 2000-2006. As your indication, we didn't study the influence of East Asia O3 pollution on US in this work, which will be the topic of our next step. The discussion in the summary part has been modified.

(4) Why the summer focus here when spring is typically understood to be the season when Asian export peaks and has maximum impact on free tropospheric ozone and trans-Pacific transport (e.g., see www.htap.org reports from 2010 or 2007)?

Response: We focused on summer because we hope to observe the contribution of lightning NOx and the influence of East Asia monsoon on that. The contribution of East Asian lightning NOx is much smaller on other seasons.

(5) Section 2.3. It seems appropriate to provide a short description of the data treatment.

Response: More description has been added.

(6) If the Jiang et al. 2014ab are not yet available in ACPD, the relevant information for which they are being cited needs to be incorporated here.

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Response: Jiang et al. 2014b has been published in ACPD and the reference list has been updated. We haven't submitted Jiang et al. 2014a yet, and thus the status has been changed to "in preparation." Fortunately, we published another paper using the same code recently. The citation has been added.

Mao, Y. H., Li, Q. B., Henze, D. K., Jiang, Z., Jones, D. B. A., Kopacz, M., He, C., Qi, L., Gao, M., Hao, W.-M., and Liou, K.-N.: Variational estimates of black carbon emissions in the western United States, Atmos. Chem. Phys. Discuss., 14, 21865-21916, doi:10.5194/acpd-14-21865-2014, 2014.

(7) The assumption of a proportional relationship between emissions and NO2 VCDs seems problematic for lightning NOx (and possibly biomass burning NOx), which would probably have longer lifetimes than NOx from the other sources.

Response: I agree with this point. Lin et al. 2012 has indicated that "any NO2 molecule originating from lightning is 1.5 times as likely to be observed by OMI than a NO2 molecule of anthropogenic origin". In this work, only the a posteriori anthropogenic NOx emission was used.

(8) Section 4.1 Is the 7 ppb TES bias specific to the region/time period being studied here? Is this determined relative to ozone sondes?

Response: It is relative to ozone sondes. However, because the World Ozone and Ultra-violet Radiation Data Centre (WOUDC) sites used in the validation (Verstraeten et al., 2013) are mainly located in Europe, and consequently it may not be an accurate evaluation for East Asia. More discussion has been added.

(9) How are the correlations in Table 1 calculated? Is the regional average first calculated for ozone and CO, and then the correlation is done for daily regional means? See also comment G3.

Response: The model values are sampled at TES measurement time and location rather than daily mean values. Description has been added in the Caption.

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(10) The conclusion of consistency between model and TES ozone-CO relationships (p19524) is subjective. It looks like in 5 out of 12 examples over E China, the model and TES suggest opposite signed changes from year to year. The abstract discusses seasonal variations, but the year-to-year changes seem more relevant given the motivation of rising emission trends.

Response: The consistency between model and TES CO is really not satisfying. We speculate a possible reason is: "the TES CO data are biased towards polluted air parcels because of its relatively low sensitivity whereas the model captures background values", which is normal in the comparison between model and measurements, particularly when the measurements are sparse. Fortunately, the consistency on O3 is much better. The abstract has been changed.

(11) In terms of the optimized emissions in Fig 1, how do we know this isn't correcting for the OH bias discussed in the text? Is there seasonal variation in anthropogenic emissions in the model?

Response: A very good question! The bias in OH has significant influence on the estimation of CO emission. In Jiang et al. 2014, we evaluated its effect in detail. We will try to improve our results with a full-chemistry inversion in next step. The seasonal variation in the anthropogenic emission is small in the a priori emission inventory, as shown in Jiang et al. 2014.

Jiang, Z., Jones, D. B. A., Henze, D., Worden, H: Sensitivity of inferred regional CO source estimates to the vertical structure in CO as observed by MOPITT, Atmos. Chem. Phys. Discuss., 14, 22939-22984, doi:10.5194/acpd-14-22939-2014, 2014.

(12) Figure 4 suggests a similar spatial pattern for lightning and anthropogenic NOx so how is this a clean separation of anthropogenic vs. natural?

Response: In the adjoint model, both transport and chemistry components are run backwards. The O3 in free troposphere will be transported to where it is produced and

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then be converted to NO2. Because anthropogenic and lightning NOx has different altitude, they are able to be distinguished.

(13) Please explain why the chemical scheme is sufficient to evaluate this partitioning on the evidence of a small relative bias (Section 4.1 p 19525). The language in the final paragraph of 4.1 is confusing.

Response: A very good question! It may not suitable to say the "the chemical scheme is sufficient" as there are still potential issues in it. The discussion has been rewritten.

(14) P19526 statement referring to Mao et al. 2013. Please comment as to what current understanding suggests and whether the effects seen in Figures 4 and 5, showing ozone decreases associated with isoprene emissions, are consistent with this understanding.

Response: Mao et al. 2013 demonstrated the sensitivity of surface O3 concentration over southeast United States on isoprene could change sign, from negative to positive, with two different isoprene scheme. It implies the sensitivity to biogenic isoprene is highly dependent on the isoprene chemistry scheme. More discussion has been added.

(15) Where did we see the anthropogenic VOC contribution from China (P19529 L 24-25)?

Response: The current model only allows us to calculate the sensitivity of O3 on anthropogenic CO and biogenic isoprene, as shown in Figure 5 and 6. The conclusion is obtained by assuming anthropogenic CO is a proxy of anthropogenic VOC and biogenic isoprene is a proxy of biogenic VOC. Thus, in Page 19526, we have: "It implies China is the major source of anthropogenic hydrocarbons and Southeast Asia is the major source of biogenic hydrocarbons". We have added more discussion about it.

Technical comments

(1) Please provide a short description of what a major quality flag equals 1 means.

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Response: We use only those data for which the "speciesretrievalqualityflag" is set to one in the data products. These data have passed all major quality flags used to assess the TES data related to chi-2 tests, biases in the radiance residuals, and residual non-linearity checks. Description has been added.

(2) For the MOPITT a priori, are the MOZART-4 fields averaged over the same 10 degree x 60 degree grid as for TES?

Response: The MOPITT a priori doesn't have the 10° latitude x 60° longitude average. The description has been changed.

(3) Section 2.2 please define what is meant by free troposphere; which retrieval levels are examined?

Response: Thanks! It has been changed.

(4) Figures 4,5,6 should state the year for the adjoint calculations (2006)?

Response: Thanks! It has been changed.

(5) Figure 2, is the model red line also smoothed with the TES AK?

Response: Yes, the model is smoothed with TES AK. The caption has been changed.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 19515, 2014.