Atmospheric Chemistry and Physics Discussions

Interactive comment on "Reconstructing ozone chemistry from Asian wildfires using models, satellite and aircraft measurements during the ARCTAS campaign" by R. Dupont et al.

Anonymous Referee #1

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Question: The structure of the paper should be improved and the different parts of the analysis consolidated more clearly. For example, the authors start by discussing flight 11 in situ and DIAL aircraft data results, then move on to giving an overview of TES and modeling tools. Model evaluation with aircraft data is very vague and spread across the manuscript. I suggest introducing all tools first, and then describe the aircraft LIDAR and measurements together with model evaluation. This way the model evaluation can be deepened by adding also modeled time series of trace species in Figures 5 and 6.

Answer: In the revised manuscript we will introduce TES, RAQMS, and then DIAL measurements (aerosol Scattering ratio and ozone) for flight 11. This will be followed by a comparison between the RAQMS Black Carbon (BC) predictions and Kondo (SP2) measurements including a curtain plot of RAQMS BC (new figure 5) which will be used along with comparisons with other insitu measurements (new figure 6) for a more through model evaluation.



New Figure 6:



Question: A restructure would also allow cutting back on the fairly high number of figures. E.g., DIAL ozone and backscatter ratio data are shown in Figures 3 and 4, and then again in Figures 9 and 10.

Answer: I removed DIAL ozone in figure 10 and DIAL ASR in figure 9 and combine the rest of the figures into a new figure 9

New Figure 9:



Page 26758, line 26:

A number of species are mentioned but only few are shown in the Figures; I suggest indicating more clearly what is shown and what not. There is no mention of measurements of trace species that are well-known fire tracers, like acetonitrile or HCN. Are measurements of these species available for Flight 11?

Answer: I mentioned the species that are or are not shown in the figures. Acetonitrile was available in the in situ data, unfortunately not during the entire part of the flight and were not relevant. There was a measurement of HCN by Paul Wennberg (wennberg.gps.caltech.edu) during the flight. It is now used to color the other insitu measurements shown in the new figure 6. Also note that BC is now presented in ng/m3 instead of #/cm3 in the ne figure 5 since we are comparing the results to RAQMS, which doesn't carry number density.

Page 26760, line 5:

Please also state values for TES CO validation results.

Answer: TES CO retrieval accuracy have been assess through comparisons to aircraft insitu measurements during INTEX-B (International Chemical Transport Experiment) 2006, AVE (Aura Validation Experiment, Houston, TX) 2004, CR-AVE (San Jose, Costa Rica) 2006, and PAVE (Polar Aura Validation Experiment) 2006 (Luo et al., 2007). The agreement between TES CO profiles and in situ data is typically within 15% and Lopez et al. (2008) reported that, in the 700-200 hPa pressure range where TES is sensitive to CO, in-situ measurements from the WB-57 aircraft agree with TES to within 5-10%. Global comparisons between MOPITT (Measurement of Pollution in The Troposphere) and TES CO measurements have been performed as well (Ho et al., 2009). The results show that for pressure layers where both instruments are most sensitive, the retrievals agree to within 10%. The global CO pattern observed by TES shows similar qualitative features to those seen by MOPITT. Comparison between TES CO data in the upper troposphere and those from the ACE instrument show an agreement of 7.4% at 316 hPa.

New References:

Ho, S., D. P. Edwards, J. C. Gille, M. Luo, G. B. Osterman, S. S. Kulawik, and H. Worden, A global comparison of carbon monoxide profiles and column amounts from Tropospheric Emission Spectrometer (TES) and Measurements of Pollution in the Troposphere (MOPITT), *J. Geophys. Res.*, *114*, D21307, doi:10.1029/2009JD012242, 2009.

Lopez, J. P., M. Luo, L. E. Christensen, M. Loewenstein, H. Jost, C. R. Webster, and G. Osterman, TES carbon monoxide validation during two AVE campaigns using the Argus and ALIAS instruments on NASA's WB-57F, *Journal of Geophysical Research*, *113*, doi:10.1029/2007JD008811, D16S47, August 15, 2008.

Luo, M., C. Rinsland, B. Fisher, G. Sachse, G. Diskin, J. Logan, H. Worden, S. Kulawik, G. Osterman, A. Eldering, R. Herman and M. Shephard, TES carbon monoxide validation with DACOM aircraft measurements during INTEX-B 2006, *J. Geophys. Res.*, 112, D24S48, doi:10.1029/2007JD008803, December 20, 2007.

Page 26760, line 25:

How where fire emissions treated in the model? Was a fire injection height considered?

Answer: Biomass burning emissions in the RAQMS model includes daily, ecosystem and severity based estimates based on Moderate Resolution Imaging Spectroradiometer (MODIS) Terra and Aqua fire detections [Al-Saadi et al, 2008]. Fire emissions are a function of the fire severity and are injected over the depth of the planetary boundary layer for all fires except severe boreal fires, in which case the fire emissions are injected to 1km above the planetary boundary layer. Fire severity is estimated using the Haines index [Haines, 1988] developed by the US Forest Service.

New references:

Al-Saadi, J. A. et al., (2008) Intercomparison of near-real-time biomass burning emissions estimates constrained by satellite fire data, J. Applied Remote Sensing, Vol. 2, 021504

Haines, D.A. 1988. A lower atmospheric severity index for wildland fire. National Weather Digest. Vol 13. No. 2:23-27.

Page 26762, line 7:

I am confused by the definition of the baseline simulation. In the model description it is stated that the current study applies assimilation of O3 and AOD satellite data in RAQMS and I assume this is considered the baseline simulation. Then, on page 26764, line 21 it is mentioned that the sensitivity simulations do not include assimilation. This implies, that when the baseline and sensitivity results are subtracted to estimate the fire influence, then

it is not only differences in fire emissions that are impacting this difference. How does this model inconsistency impact the results? And why was a baseline simulation not conducted without assimilation to allow consistency between all model runs?

Answer: Comparisons of RAQMS with observations include assimilation of O3 and AOD satellite data. On the other end, the Baseline simulation used in the sensitivity study does not include assimilation. As a matter of fact, only differences in fire emissions impact the differences between the baseline and sensitivity tests.

Question: I also suggest moving the description of the sensitivity simulations earlier to Section 2.4 (model overview).

Answer: I moved section 3.1 to RAQMS overview in section 2.3 (previously 2.4) as stated.

Section 3.2:

I suggest moving this section to the discussion part of the paper (Section 4) after the plume evolutions have been discussed.

Answer: I moved section 3.2 to section 4 as stated.

Section 3.3.1:

The model performance for this plume analysis is in my opinion rather poor. The authors do not state any possible reasons for the high CO - low O3 bias in the model and in what way this impacts their conclusions.

Answer: The new Section 3.3.1. will include a more complete discussion about CO and O3 bias between RAQMS and TES. The initial comparison between TES and RAQMS with the TES averaging kernel applied (left side of figure 11a) shows good agreement in maximum CO (which is associated with the wild-fire plume) but RAQMS shows larger minimum CO (160ppbv vs 110ppbv). This is due to overestimates in RAQMS background CO mixing ratios that increase with latitude in the northern hemisphere (also evident in the comparison with insitu measurements on 04/19 (see new figure 6). RAQMS ozone initially (04/07) ranges from 45-65ppbv which is in good agreement with TES except for a few points with O3 between 70 and 90 ppbv (also evident on 04/14, figure 11b). These retrieved ozone enhancements are found in the upper troposphere in the TES retrieval (see below) and not associated with the Kazakhstan wildfire plume and indicate underestimates in stratospheric-tropospheric exchange in RAQMS relative to the TES retrievals. The magnitude of the wild-fire CO enhancement for this plume was initially \sim 500ppbv on 04/07 (see lower right in figure 11a) and diminishes to slightly larger than 15 ppbv by 04/18. As the plume diminishes the overestimates in RAQMS background CO account for most of the differences found in figures 11 b and c.

Page 26767, line 15:

The last 2 sentences of this section do not make sense. If there is significant cloudiness in an area, should there be even any TES retrievals?

Answer: TES averaging kernels are reduced with significant cloudiness and then the retrieval reports the a priori.

Page 26768, line 27:

Specifying the latitude where this happens would make the localization in the graphs easier.

Answer: I've added the location of the plume in the text.

Section Discussion:

It would be beneficial to provide in this Section also a brief summary and comparison of the TES/RAQMS/aircraft derived ozone enhancements (dO3/dCO) before the modeled enhancements are extrapolated to a larger picture.

Answer: This section will be improved in the next version of the paper. I will do a quick summary of TES/RAQMS/aircraft ozone enhancements.

Figures 4 and 10:

Why are there gaps in the ozone curtain in Figure 10 but not in Figure 4?

Answer: I removed DIAL ozone curtain from figure 10 and referred to figure 4 for comparison. Figure 10 was plotted using a first version of the data with a coarser resolution and without interpolation across the near field region.