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Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #2

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This paper discusses the transport and chemical evolution of Asian Biomass Burning (BB) plumes across the Pacific. It makes use of aircraft measurement from the ARCTAS campaign (both in-situ and lidar measurement), of CO and ozone measurement profiles from the TES satellite, and from RAQMS model simulations. Using trajectory analyse, authors are able to follow plume transport and to analyse TES measurement and RAQMS simulations at different location of the plume (close and far from emissions) up to the intersect with the ARCTAS aircraft, along the West American coast.

The manuscript is well written, but not always well organized. The subject of the paper is of scientific interest as transport of biomass burning plume and production of ozone

in those plumes are not fully understand yet. Modeling vertical transport and chemical history of biomass burning plumes is a particularly difficult task and interesting question. One very interesting aspect of this study is the use of several measurements, from in-situ and remote sensing platforms, together with model simulations with the aim to follow plume evolution.

However I found the analyse of the comparison between model and measurement rather poor and the discussion part too extensive in regards to this analyse. Comparisons with in-situ data are not fully performed and are treated all along the paper without real coherence. Strong discrepancies between in-situ measurement and model are not really highlighted and reasons for these discrepancies not enough discussed (why is the highly polluted BB plume observed by aircraft not well simulated by RAQMS and seen by TES ? In particular, why is ozone not strongly produced in that plume according to the model, when observations show strong ozone production ?). More generally, authors present interesting comparisons between TES and RAQMS model but, as for in-situ data, there is no real analyse of discrepancies. There is also no analyse of the reason why model simulate strong ozone production in Thailand plume and not in Kasakhstan one.

I think more work should be done before the manuscript is published. Authors should concentrate on a more systematic analyse of data/model comparison. Authors should also put their work into context: they should show what this work brings compared to older studies on BB plumes transport over the pacific (for example during ITCT 2K2 or INTEX-B).

The following text gives more details and suggestions:

In the abstract: It may be interesting to give mean plume altitudes.

Section 2.2: using aerosol DIAL measurement, authors identified 2 BB plumes: one in the middle/low troposphere, and the other in the upper troposphere. According to text and Figures 3 and 4, plumes are first identified using aerosol enhancement detected by DIAL measurement. For the middle tropospheric plume, in-situ data confirm the BB origin (strong enhancement in CO and BC levels). For the upper tropospheric plume, it is not clear why authors reduced the plume to the signal observed between 43°N and 55°N, and not to signal observed between 36°N to 40°N at about the same altitude, that shows similar enhancement and for which in-situ data exist (with enhancement in CO and PAN). Figure 10 showing RACMS simulations also suggest that aircraft may have measured the upper tropospheric plume between 36°N and 40°N (23.00 UTC to 24.00 UTC), even if this part of the plume seems more diluted that the one mentioned in the text. I suggest to take advantage of these in-situ measurements of the upper tropospheric plume, or explain why this signal cannot be considered as BB plume. Is there any measurement of acetonitrile ? As it is a good BB tracer, this could help to distinguish between BB and non BB plumes.

section 2.2.2 at last paragraph: "We use these aircraft data along with forward wildfire trajectories... and explain the ozone distributions observed during NASA/DC8 flight 11 on the 19 April 2008". If I understand well, "these aircraft data" and " NASA/DC8 flight 11 on the 19 April 2008" are the same flight. It is a bit confusing, why not use the same words (or "these aircraft" or "NASA/DC8 flight..")?

Section 2.4: there is no information on how BB emissions are simulated in the RAQMS model. Which data are used for fire detections, which emission factors are used, is there any fire injection high ? These data are really important in order to evaluate BB simulations from the RAQMS model.

Section 3.1 and Figure8: I It is difficult to see trajectories from the figure, trajectories may be enlarged. Authors should also mentioned how forward trajectories were selected, as more than thousands of forward trajectories were probably initiated from the fire emissions (maybe backward trajectories were used first ?). Also, there is no mentioned of "NOy emissions" either in the text or in the figure caption. It should be explain somewhere. It would also be interesting to see the altitude of this forward trajectories in order to see when occurs the uplift of air masses.

It is not clear why authors detailed RAQMS comparison with DIAL data in this section. As suggested by Reviewer 1, a reorganisation of the paper would help the reader, with a section dedicated to comparison between RAQMS and aircraft data (in-situ and DIAL measurements). For example, Figure 12 (showing comparison between RAQMS and in-situ ozone/CO correlation) would fit in this section (after axes are changed: it's better to keep CO in abscissa and ozone in ordinate as in the other figures). In this new section, authors should emphasis the large differences between ozone and CO simulated and measured by the aircraft in the middle/low troposphere. As shown in Figure 12, the model is not able to simulate observed ozone and CO range. Authors should suggest explanation for these large discrepancies. In the paper, some explanations are given in the conclusion but it should be more discussed in the text himself. Is it possible that RAQMS advected the plume more to the north or to the south ? Do authors look at RAQMS simulation in the vicinity of the aircraft (and not directly along the aircraft)?

It should also be noticed that the O3/CO correlation measured by the aircraft has a positive slope in the plume (i.e for CO > 150 ppbv for example), showing ozone production along the transport (Parrish et al., 1993, Pfister 2006 etc ..). RAQMS simulation does not show any positive tendencies for this O3/CO correlation. So even if the plume in RAQMS is too diluted due to its numerical resolution, the question of the ozone production in the plume should be discussed. More generally, the O3/CO correlations observed or simulated can be used to have more informations on ozone production. For example, when looking at Figure 13, it seems that RAQMS simulate an increase in O3/CO slope in the plume (CO>150 ppbv for example) during the transport, suggesting ozone production en route (mean ozone in the plume also seems to increase).

Section 3.3.1: "Comparison between the TES and the adjusted RAQMS model for these points indicates that RAQMS has lower background ozone in this region relative to TES". RAQMS has also higher CO background. Authors should give an explanation for this lower ozone background and higher CO background that is found in Figure 11 a) b) and c), for example it may be due to stratospheric intrusion badly represented or wrong concentrations (CO overestimated and ozone underestimated) in the upper troposphere in the RAQMS model. More generally in the paper, authors should try to find explanations for discrepancies found between model and measurement and not only described these comparisons.

“At this stage, ozone concentrations in the plume are up to 60 ppbv, 10 ppbv over background level”. How are background level estimated ?

“Both TES and the adjusted RAQMS model show maximum CO concentrations (around 225 ppbv) and ozone concentrations.” The sentence is not clear, you could maybe add ’show similar maximum ...’

“RAQMS underestimates maximum in situ CO concentrations within the wildfire plume”: ozone concentrations are also underestimated.

Section 3.3.2 The paragraph from “Figure13a, b and c” to “adjusted RAQMS model, and TES” may be removed or reduced as the same kind of figures were explained in the previous paragraph. Reference to Figure 14 do not correspond to the figure (left, right etc ...). This may be due do a reorganization of the figure by the ACP editor team. In the on-line version, Figure 14 groups 6 large figures. It is disproportionated regards to the comment on this figure (about 7 lines). This figure is mainly used to show the presence of upper-tropospheric/low-stratospheric air masses in proximity of the plume and should be or reduced or more commented.

“Along the flight track, TES and the adjusted RAQMS model ...”, are TES data really interpolated along the flight track ? Otherwise, it’s maybe better to write “at flight location” or a similar sentence.

Section 4: Comparisons between RAQMS simulations and measurement are not really good for the Kazakhstan fire, especially for dO3/dCO comparison with in-situ aircraft measurement. Therefore, extrapolation of the model result should be done with caution. As suggested by the other reviewer, a comparison of measured and modeled

ozone enhancement (dO3/dCO) would be useful, together with a rough estimation of model uncertainties.

Figures: There is a lot of figures. Do you really need to show both Fig 1 and 7 ? Figure 6 : it is difficult to read green data. Figure 11 and 13: sometimes, CO scale is not readable (11b for TES and RAQMS retrieval and 13a for RAQMS). Figure 12 : as mentioned in the text, it would be coherent with the rest of the figures and with older studies to exchange abscissa and ordinate. Figure14 : captions and figures are not coherent.

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