**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 #2**

Received and published: 12 January 2011

Question: However I found the analysis of the comparison between model and measurement rather poor and the discussion part too extensive in regards to this analyze. 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?).

Answer: The new figure 5 and figure 6 ( see response to reviewer #1) show detailed comparisons between the RAQMS analyses and insitu measurements from the DC8 on 04/19. These comparisons show that the elevated CO, BC, NOx, SO2, and PAN due to BB (high HCN) observed by the aircraft is underestimated at 20-21 UTC because RAQMS did not capture the narrow (2km thick) elevated plume found at 4km in the DIAL aerosol scattering ratio and sampled by the DC8 at this time. Instead, RAQMS shows a broader plume that is just grazed by the aircraft. This broader plume was sampled again at 23 UTC, where RAQMS shows somewhat better agreement with the in-situ measurements but is still low. These underestimates are a consequence of diffusive transport associated with the relatively coarse horizontal (2x2 degrees) and vertical (0.5-1km in the free troposphere) resolution of the RAQMS model. RAQMS shows no NOx enhancements in the plume at 20-21 UTC and consequently underestimates ozone production.

Question: More generally, authors present interesting comparisons between TES and RAQMS model but, as for in-situ data, there is no real analyze of discrepancies. There is also no analyze of the reason why model simulate strong ozone production in Thailand plume and not in Kazakhstan one.
I think more work should be done before the manuscript is published. Authors should concentrate on a more systematic analyze 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:

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

Answer: I mentioned the plumes height in the abstract.

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 taking advantage of these in situ measurements of the upper tropospheric plume, or explain why this signal cannot be considered as BB plume.

Answer: The second period of significant HCN, CO, PAN, BC, SO2, and NOx enhancement occurs at 23 UTC and is associated with the lower tropospheric plume. The upper tropospheric plume sampled by the DC8 between 36-40N shows high O3, and NOx but low CO, PAN, HCN, and BC and is not likely to be of BB origin. Note from figure 8 a and b that the Thailand and Kazakhstan plumes were transported to the North West of 36 - 40N. Note from new Figure 9 (see response to reviewer #1) that the core of the Thailand plume is aloft and the DC8 only grazed the southern part of this plume.

Question: Is there any measurement of acetonitrile? As it is a good BB tracer, this could help to distinguish between BB and non-BB plumes.

Answer: There was in situ measurement of acetonitrile that, I agree, would have been really useful. Unfortunately, data are only available for the first part of the flight until 21 UTC but shows high levels of acetonitrile. I will mention those levels in the text but will not show them in a figure. However, HCN, also a good BB tracer, has been added to the analysis as a tracer of BB plume in the new figure 6 (see response to reviewer #1).

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..")?

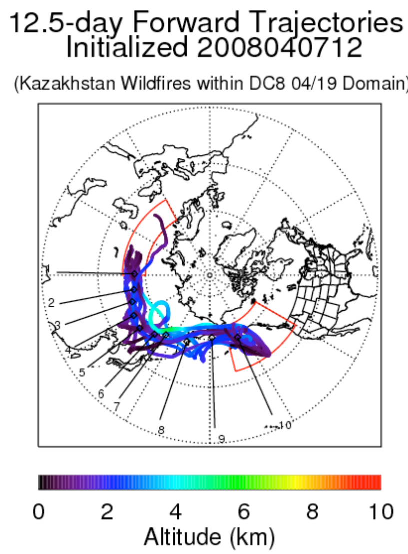
Answer: I replaced NASA/DC8 by aircraft.

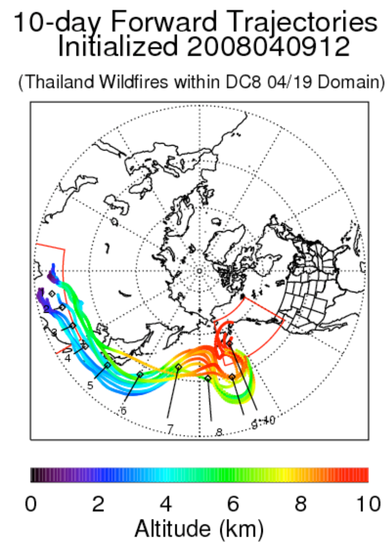
Section 2.4: There is no information on how BB emissions are simulated in the RAQMS model. Which data are used for fire detections, which emission factors are used, is there any fire injection high? These data are really important in order to evaluate BB simulations from the RAQMS model.

Answer: See response to reviewer #1 regarding Page 26760, line 25.

Section 3.1 and Figure 8: It is difficult to see trajectories from the figures, they may be enlarged.

Answer: Figures 8a and 8b have been enlarged and the color-coding shows altitude along the trajectories instead of NOy emissions




New Figures 8 a and b

Question: 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?).

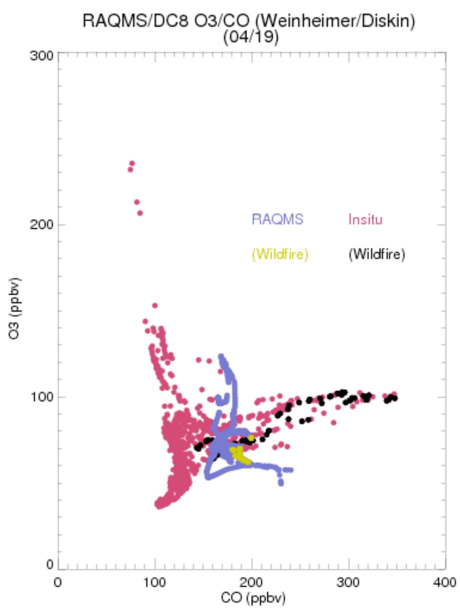
Answer: The subset of forward trajectories was selected by where they began (either Thailand or Kazakhstan) and where they ended up on 04/19 (near the DC8). These boxes are now indicated in the new figures 8 a and b above.

Question: 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 these forward trajectories in order to see when occurs the uplift of air masses.

Answer: NOy emissions have been removed and replaced by the altitude of the forward trajectories which clearly shows up when the uplift occurs.

Question: It is not clear why authors detailed RAQMS comparison with DIAL data in this section. As suggested by Reviewer 1, a reorganization 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)?

Answer: We have added the RAQMS analyses to the in-situ timeseries and will more thoroughly discuss the reasons for the discrepancies (see response to reviewer 1). We have updated figure 12 (blow) to be consistent with figures and will move this figure forward to the discussion of the in-situ vs RAQMS. We have also improved the process for determining whether the DC8 was sampling BB plume by using an HCN threshold (1500pptv) to identify BB plumes (new figure 6). This was accomplished by interpolating the 1-sec O3 and CO measurements to the ~15 sec HCN measurements. The new figure 12 (below) shows the comparison between the DC8 and RAQMS O3/CO scatterplot within the BB plume (HCN>1500ppbv)




New figure 12

Question: 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 information 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).

Answer: Positive slopes in the O3/CO correlation and chemical production will be discussed in more details in the updated version of the manuscript.

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.

Answer: We will take a better care on studying the discrepancies between TES and RAQMS in the updated version of the manuscript.

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

Answer: Background levels are estimated based on monthly mean TES O3 observations in the troposphere.

Question: "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...'

Answer: I've rephrased the sentence.

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

Answer: The O3 underestimation is stated at the end of the sentence.

Section 3.3.2 The paragraph from "Figure13 a, 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.

Answer: I've reduced the paragraph and referred to the previous section for description of the figures.

Question: 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 disproportionate 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.

Answer: I reduced the number of figures (2 instead of 6), kept the figure that show dO3 and dCO due to the fires (originally figure e and f) and applied the modification due to ACP figure reorganization.

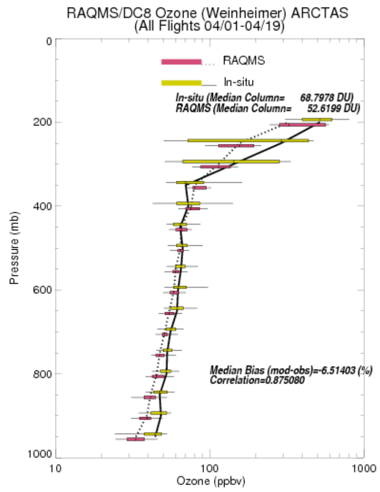
Question: “Along the flight track, TES and the adjusted RAQMS model...”, are TES data really interpolated along the flight track ? Otherwise, it’s maybe better to write “at flight location” or a similar sentence.

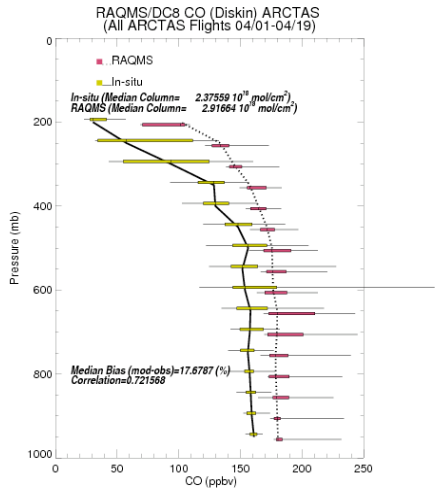
Answer: TES data are not interpolated along the flight track. I substituted “flight track” by “flight location”.

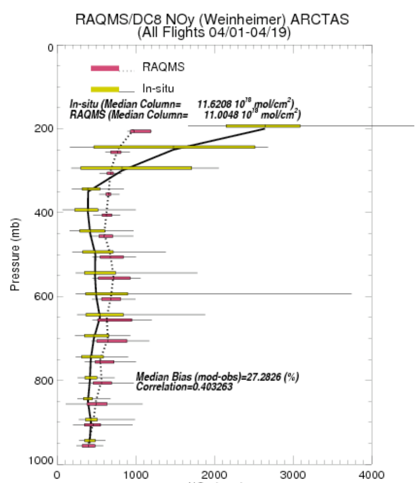
Section 4: Comparisons between RAQMS simulations and measurement are not really good for the Kazakhstan fire, especially for dO3/dCO comparison with in-situ aircraft measurement. Therefore, extrapolation of the model result should be done with caution. As suggested by the other reviewer, a comparison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

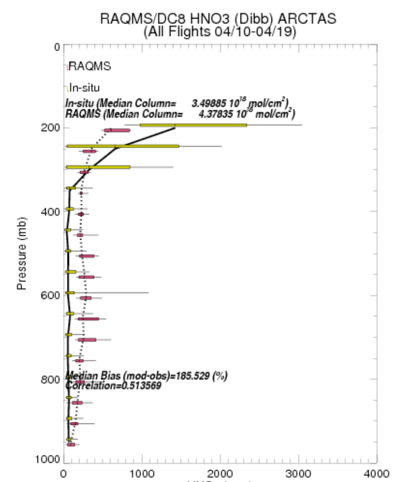
Answer: I have moved section 3.2. discussing the comparison of measured and modeled ozone enhancement (dO3/dCO) to section 4. Statistical comparisons with all DC8 insitu measurements between 04/01 and 04/19 have been used to provide a better estimate of model uncertainties (see figures below). Overall, RAQMS ozone analyses is very well correlated with the insitu measurements (r=0.875) but shows a 6.5% median low bias with largest biases above 300 mbar associated with underestimates of lower stratospheric ozone. RAQMS CO is well correlated (r=0.72) with insitu measurements but has a 17.67% median high bias associated with a 20ppbv overestimate in background mixing ratios. Comparisons between RAQMS NOy and HNO3 and in-situ measurements result in somewhat lower correlations (r=0.4 and 0.51, respectively) and show that RAQMS underestimates stratospheric HNO3 (and thus NOy) above 300 mbar and overestimates tropospheric HNO3 (and thus NOy) below 300 mbar. Comparisons of RAQMS NOx and PAN with insitu measurements shows lower correlations (r=0.14 and 0.31, respectively) due to the shorter lifetimes of these species and shows that RAQMS systematically underestimates NOx by 46.7% and overestimates PAN by 54.9% with largest discrepancies found above 300 mbar associated with underestimates of stratospheric influences.

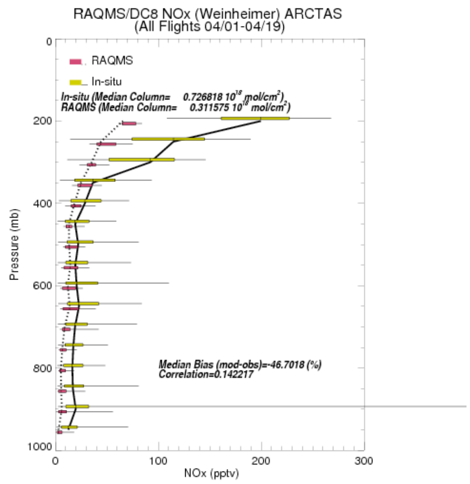
The systematic underestimate of stratospheric influences and overestimate in background CO relative to the insitu measurements is consistent with the underestimate of ozone and overestimate of CO relative to TES for the Kazakastan plume. Underestimates in NOx and overestimates in PAN are consistent with reduced ozone production efficiency and, combined with diffusive transport of the narrow BB plume observed by the DC8 on 04/19, account for the underestimates of dO3/dCO relative to insitu measurements.

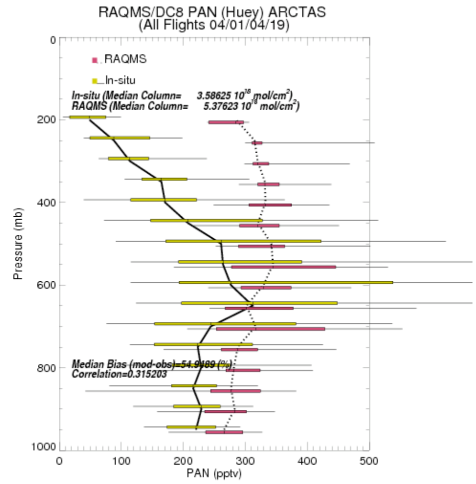




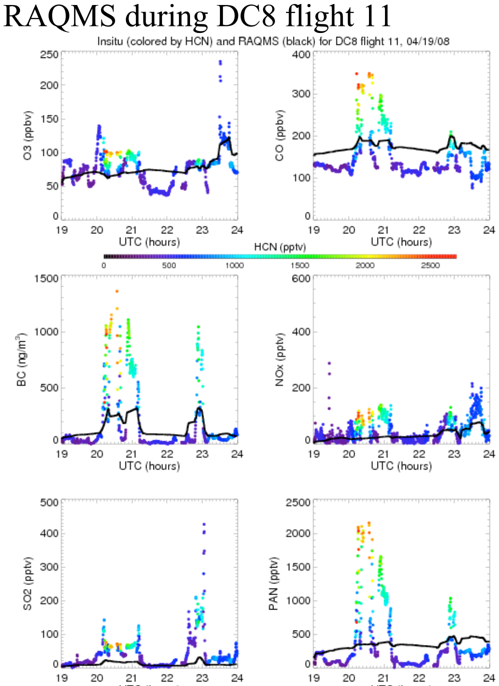




Question: There is a lot of figures. Do you really need to show both Fig 1 and 7?

Answer: I think figure 1 is important to help the reader localize the fires. Figure 7 will be deleted.

Question: Figure 6: it is difficult to read green data.

Answer: Figure 6 has been updated to a 6 panels figure (see below) comparing in-situ and RAQMS during DC8 flight 11

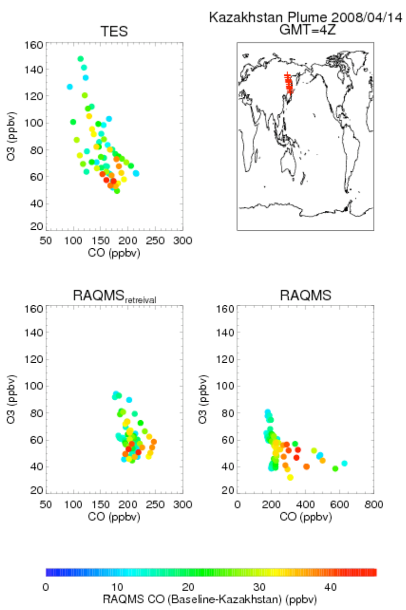



New figure 6

Figure 11 and 13: Sometimes, CO scale is not readable (11b for TES and RAQMS retrieval and 13a for RAQMS).

Answer: We improved the readability of the figures (reduce numbers in abscissa: see above)

New Figure 11b




Question: 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.

Answer: We switched abscissa and coordinate to fit the rest of the figures template (see previous answer to reviewer #2 above)

Question: Figure14: captions and figures are not coherent.

Answer: As mentioned above, I reduced the number of figures (2 instead of 6) and applied the modification due to ACP figure reorganization.