Atmos. Chem. Phys. Discuss., 9, C2942–C2952, 2009
www.atmos-chem-phys-discuss.net/9/C2942/2009/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evaluating simulated primary anthropogenic and biomass burning organic aerosols during MILAGRO: implications for assessing treatments of secondary organic aerosols" *by* J. D. Fast et al.

J. Fast

jerome.fast@pnl.gov

Received and published: 17 July 2009

The authors would like to thank the reviewers for their thoughtful comments and suggestions. Our responses are given in the order they were listed, and we have included the comments in our response for completeness. The major changes include the removal of Section 4.5 on TOOC since we agree that a more comprehensive analysis using other sites is needed, the addition of source attribution for simulated POA that changes the discussion of biomass burning sources of organic matter, and additional discussion throughout the text regarding details of the model configuration. Many of

the figures have also been altered. We believe that the revised paper has responded to all of the reviewer's comments.

Comment: On the possible overestimate of emissions from large fires. When flying directly downwind of fires the aircraft evidently measured less POA than the model predicted and the conclusion is that the POA emissions may have been overestimated from "some large fires." It seems it may be possible to eliminate some specific causes of overestimates rather than concluding with that general of a statement. To explore this; a true overestimate could result in at least three ways:

(1) The assumption of too high a particle production per unit mass of fuel burned. This seems unlikely to occur since Yokelson et al. (2007) measured fire-averaged particle emission factors that were similar to the literature average for the type of fires most frequently observed in the MC-area. It is my understanding that values very close to these were also used in the fire predictions used in this study. In fact, wasn't the real PM/CO ratio measured precisely while flying in the large plumes?

(2) The assumption of too large a total fuel consumption or burning rate. Most likely the fire emissions model assumes that most of a pixel burns if it registers as a hotspot. This may often be the case, but at a given time when the aircraft samples in the plume only a small portion of the pixel is likely to be actively burning.

(3) As Figure 3 in Yokelson et al. (2007) shows, the particle emissions for a fire depend to some extent on the relative amounts of flaming and smoldering combustion, which can change during a fire.

The disagreement when flying downwind of fires could also occur due to incorrect mixing or plume rise. If mixing was the problem then other fire emissions would also be overestimated by the model. This seems to be the case on page 4823 lines 1-2 where it is noted that CO is also overestimated downwind of large fires. It was not clear to me if this was the case with other fire emissions or not (possibly for TOOC (vide infra)). If mixing can not be eliminated as a source of error it may help to know that the March

ACPD

9, C2942–C2952, 2009

fires were quite small and the plumes seemed to have much less buoyancy than is commonly observed. Indeed, near-horizontal and even downslope flow was commonly observed for fresh fire emissions released in the middle of the local afternoon. This type of flow could be quite difficult to simulate in any model. I bring this up mainly because it would ultimately be useful to know if the emission ratios for the fires probably are "OK."

Response: The reasons for an uncertainties in biomass burning estimates given by the reviewer all certainly possible. Unfortunately, our modeling study was not designed to narrow down the source of the errors to a specific cause. Uncertainties in boundary layer vertical mixing and treating plume rise contribute to our model predictions that make it problematic to identify the specific uncertainties associated with fire loading. A modeling study that focuses on specific fires (at smaller grid spacings than in this study) and has sufficient meteorological, chemical, and aerosol measurements would be more appropriate when assessing biomass burning estimates. Instead, this study uses the best biomass burning estimates available at face value. We have, however, modified Section 3.3 to include additional sources of uncertainties than those we already listed. The reviewer also seems to be noting the behavior of specific fires based on visual inspection. If this type of information was quantified in some way, or available via movies, it would be very helpful to examine simulated fire plume behavior. We have also modified some of the text regarding large fires, based on other comments made by the reviewers that have changed the figures.

Comment: On separating HOA and POA and BBOA. Just a word of caution that freshly emitted BB OA has fairly high oxygenated content. Moffet et al and Crounse et al found that a significant part of the OA in the ambient urban air in MC was "fresh BB aerosol." Does this effect the PMF separation for the AMS particle types? If not, the reason could be provided in a sentence near the beginning of the paper. A related general topic is that the AMS separates fresh and aged aerosol in a pure urban environment as HOA and OOA respectively. It wasn't clear to me where the fresh and aged components of BB aerosol show up. Are they both in BBOA or is some aged BB aerosol showing up

ACPD

9, C2942–C2952, 2009

in OOA? This could be clarified at the outset on page 4809.

Moffet, R. C., de Foy, B., Molina, L. T., Molina, M. J., and Prather, K. A.: Measurement of ambient aerosols in northern Mexico City by single particle mass spectrometry, Atmos. Chem. Phys., 8, 4499-4516, 2008.

Crounse, J. D., DeCarlo, P. F., Blake, D. R., Emmons, L. K., Campos, T. L., Apel, E. C., Clarke, A. D., Weinheimer, A. J., McCabe, D. C., Yokelson, R. J., Jimenez, J. L., and Wennberg, P. O.: Biomass burning and urban air pollution over the Central Mexican Plateau, Atmos. Chem. Phys. Discuss., 9, 2699-2734, 2009.

Response: A sentence has been added to this paragraph stating that BBOA is composed mostly of primary emissions. SOA formed from biomass burning emissions appears as OOA (Grieshop et al., 2009).

Comment: Finally, the possibility of trash-burning was raised by Referee #1. There are now emission factors for trash burning in the MCMA measured by Christian et al. (2009). The EF may have limited value in the context of this study until an inventory of trash burning is completed. However, an extremely rough estimate suggested that trash burning could account for _ one-third of the PM in the MCMA. Also, a personal communication from Jose Jimenez suggested that there may now be AMS spectra of trash burning emissions (high HCl, high Sb?). It may be possible for the revised version of the paper to include a few sentences that assess the possible affects of this source on the model measurement comparison. In addition, the same paper notes that many brick-making kilns were located near the T1 site. Also, possibly worth mentioning.

Christian, T. J., Yokelson, R. J., Cárdenas, B., Molina, L. T., Engling, G., and Hsu, S.-C.: Trace gas and particle emissions from domestic and industrial biofuel use and garbage burning in central Mexico, Atmos. Chem. Phys. Discuss., 9, 10101-10152, 2009.

Response: We have added a paragraph at the end of Section 3.3 to indicate that trash burning is a possible source of PM. As indicated in the new text, it is probably

ACPD
9, C2942–C2952, 2009

premature at this stage to include them in model simulations.

Comment: On POA volatility. It seems if the POA is volatile then the assumption that it is not would cancel an underestimate in the emissions inventory to some extent. Is it possible to make a simple estimate of what percent of the POA might be volatile and how that affects the assessment of the accuracy of the emission inventories?

Response: Not at present. There are a few new papers describing simple relationships that could be used to test volatility, but there are likely more studies being performed that may or may not arrive at the same conclusions. One needs to wait until a better consensus on this issue before implementing volatility representations in 3-D models when assessing emission inventories.

Comment: On VOC and PM emissions used in this work. Earlier studies by West et al seemed to indicate that the MCMAEI underestimated VOC emissions. In this work the model overpredicted the VOC. It should be very clear if both models used the same EI or if the VOC used in this work were adjusted upward and perhaps adjusted upward too much? It seems on page 4815 that the VOC in the inventory were adjusted upwards by 65%. However on page 4816 the argument is given that the PM emissions may be 30-75% too low, but it is not clear if they were adjusted upwards. Perhaps Table 3 should have rows that list the official values and the values used in this study side-by-side. Also the slopes in Figure 2 should be given in Figure 2 or the caption or the text on page 4816 so the reader can quickly check if they seem reasonable.

Response: We did not adjust the PM emissions – VOC emissions were the only emissions modified by Lei et al. The values in Table 3 for the 2002 inventory are from the original inventory, not the modified one by Let et al. We added some text in the Table to avoid confusion and did not add another column that would just reflect the one value that was adjusted (the total VOC for 2002 inventory). The slope of POA to CO was already listed on Page 4816. To clarify issues raised by all the reviewers associated with the emissions, the material in Section 3.3 has been reworded to more clearly indi-

ACPD

9, C2942–C2952, 2009

cate 1) 2002 MCMA inventory included particulate speciation and this was used in the model runs, 2) the “average speciation” from the 2002 MCMA inventory was applied to NEI 1999 PM2.5 estimates, 3) VOC changes were made by Lei et al. and that gas phase emissions are either from 2002 MCMA as modified by Lei et al. or from 1999 NEI, and 4) VOC speciation and adjustment profiles were performed by Lei et al.

Comment: General question on the nocturnal boundary layer. Is there a reasonably simple way to force the nocturnal boundary layer depth to agree better with the measurements and could it be used in future iterations of the model? Response: One could force the PBL depths to match the observations for a particular grid cell, but there is no way to extrapolate observed PBL depths far away from the sparse observations (3 sites). Also forcing the PBL depth in this manner could produce unexpected numerical problems because of inconsistencies with other properties. Increasing the weights in the data assimilation scheme could improve the vertical temperature structure, but the artificial forcing term can also produce numerical instability when the data assimilation tendency becomes larger than the other tendencies in the model.

Comment: Page 4823, lines 14 and 22: I think the Figure referred to is Figure 8 and not 6 in both cases?

Response: Yes. The reference to the figure has been corrected.

Comment: General question on background CO. It has been my experience that background CO is significantly higher in the boundary layer than in the free troposphere. Is this the case in the model?

Response: Yes, the background CO values aloft in the free atmosphere are lower than those near the surface.

Comment: P4826, L18-19 A sentence on why these two days were chosen may be useful here. Also quoting all times in LT instead of UTC allows a faster read of paper.

Response: The equivalent LT values have been added to the text in parenthesis, but

ACPD

9, C2942–C2952, 2009

we have left the figures in UTC. UTC is the standard time convention for meteorological journal articles that is used to avoid confusing local time with local daylight time.

Comment: P4827, L19 Missing “of” before “HOA”?

Response: Added “of” where indicated.

Comment: P4827, L29 Missing “a” before “mountain”?

Response: Added a missing word: used “the” instead of “a”.

Comment: P4828, L10-11 “compounds” usually used for gases? Another word better?

Response: Changed “compounds” to “components” here an elsewhere.

Comment: P4828, L23-25 Would smoothing of the topography also be a factor at Paso de Cortes?

Response: Yes, but not as significant as at Pico Tres Padres. The ridge along the east side of the basin is a much larger feature that can be resolved by the model.

Comment: P4829, L5 Here the observation of Christian et al of a large number of brick kilns near T1 may be relevant?

Response: It is likely that brick kilns are not the only uncertainty not accounted for the by emission inventories. We have added a reference to the Christian et al. paper, however.

Comment: P4829, L24 Here it would be good to remind the reader if the original or a scaled inventory was used.

Response: We have stated in the emissions description section that PM emissions were not modified, so adding a word, such as “original”, might imply that we did revise the PM emissions. Therefore, the text remains the same.

Comment: P4830, L1-6 Would comparing integrals over flight periods be a way to separate out errors in transport?

ACPD

9, C2942–C2952, 2009

Response: Only if the transport errors are reasonably small. If the simulated plume just misses the aircraft flight path, other statistics would be needed to quantify errors in transport. The percentile plots, which are already employed, are one means of relaxing the stringent point-by-point comparison between observed and predicted quantities. Instead of introducing a new statistics, we have chosen to retain the time series to show both the observed and predicted spatial variability in CO and organic aerosols.

Comment: P4830, L21-24 Re missing BB sources in urban area could include those studied by Christian et al as well as grass fires.

Response: The scale of plot has now changed with the revised figure to better show BBOA and the POA resulting from biomass burning emissions in the model, as suggested by reviewer #I. These two components are actually closer than stated in the paper because the values were misread. The text has now been corrected to reflect the new figure.

Comment: P4831, L7-14 Again, were other fire emissions overestimated or just POA?

Response: This text has been changed significantly to reflect the new figure requested by Reviewer #1.

Comment: P4832, L7-9 I'm a little uncomfortable with how this is expressed. It seems to say that deleting large fires makes for better agreement. Yet we know, and it is stated earlier in the paper, that the large fires are already an underestimate of the total BB. In general, perhaps the problem with large fires can be narrowed down and described a little more specifically as well as addressing the implications.

Response: Since Reviewer #1 suggesting performing an analysis of anthropogenic and biomass burning POA separately, the discussion of this text has changed. We no longer segregate periods with large fires.

Comment: P4833, L5 Missing "of" before "predicted"?

Response: The section on TOOC has been removed, so this text is no longer in the

ACPD

9, C2942–C2952, 2009

paper.

Comment: P4833, L6-7 If TOOC is too high in some BB plumes along with PM, does this support the idea that incorrect mixing is the most likely cause of model overpredictions?

Response: The TOOC analysis has been removed from the paper based on other reviewers comments, so this text is no longer in the final version of the paper.

Comment: P4833 L19, P4834 L2-3, and P4834 L13-15 It seems that the aromatics are simulated well and they are known "high-yield" SOA precursors. Thus, the argument that part of the VOC disagreement is caused by hydrocarbons converting to PM doesn't seem strong. Is it also possible that the MCAMAEI VOC were adjusted upward too much? Also oxygenates could also decrease due to SOA since glyoxal is an oxygenate included in the model and known to be an important SOA precursor.

Response: As with the previous comment, the TOOC analysis has been removed from the paper based on other reviewers comments, so this text is no longer in the final version of the paper.

Comment: P4834, L20 No "d" on "simulate"

Response: The section on TOOC has been removed, so this text is no longer in the paper.

Comment: P4835, L13 The authors are dealing with complex terrain in simulating the meteorology and the difficulty is described well in the paper. May be worth noting if there is a "distance" corresponding to a drop-off in accuracy for a 3 km grid (if such a number exists)? In any case, could it be worth noting that the met problems are seemingly confined to small-scale local variation?

Response: A sentence has been added to the text regarding errors in the small-scale circulations.

ACPD

9, C2942–C2952, 2009

Comment: P4836, L3 Replace “predicted reasonably well” with “reasonably good”? I.E don’t need “predicted” twice.

Response: Since the TOOC analysis with the VOC evaluation at T1 was removed, this sentence has been removed as well.

Comment: P4836, L26 Replace “that can” with “and”? Also, probably they were small grass fires that may have been abundant since only one small one was spotted from the air just before landing in Toluca.

Response: Replaced as suggested. Indeed the grass fires were likely intermittent and difficult to quantify, so their relatively importance in the sentence has been altered.

Comment: P4837, L1-2 Interesting that the horizontal mixing of smoke plumes is probably too fast as this would ostensibly cause the model to underpredict the concentrations. Thus if mixing is causing an overestimate it is most likely incorrect plume rise. Thus, it would be worthwhile to note how plume rise was handled.

Response: Horizontal mixing and vertical mixing are two separate meteorological issues, and the text here is describing only the horizontal mixing issue. Horizontal mixing in the model would tend to reduce the peak smoke plume (and thus should be lower than observed), it will spread the mass over a wider area so that smoke would be overestimated surrounding the plume centerline. We have added additional material on the biomass burning estimates, including plume rise, in Section 3.3.

Comment: P4838, L6 Probably better as “were compared with” I agree with Referee #1 that this should probably be mentioned earlier.

Response: As suggested by Reviewer #1, we have added information on the model size bins earlier in the model description section.

Comment: Figures: General, the figures are great, but very difficult to read in the printed version. To some extent, readers will have to expand them on computer to get a decent look.

ACPD

9, C2942–C2952, 2009

Response: We agree. Some of the revised figures have been expanded to make the captions more legible and multiple panels have been used to reduce too many lines cluttering on plot.

Comment: Figure 13 The data seems to suggest that BB made a significantly larger contribution at T1, which may be useful in the discussion.

Response: A sentence regarding the biomass burning fraction is now included in the discussion of Fig. 14 (which is the new version of Fig. 13).

Comment: Figure 18 Interesting that there are no alkynes in the model since it is widely used as an urban tracer. Also, were backgrounds subtracted from the species other than CO?

Response: The section on TOOC has been removed, so this text is no longer in the paper. We did not remove background values for the other species, but they were all very low and concentrations were largely due to local emissions.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 4805, 2009.