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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.

Anonymous Referee #2

Received and published: 10 May 2009

I am in general agreement with Referee #1 and the comment posted by Thomas Karl. I'll make some additional suggestions and point out a few possible typos.

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:

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(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 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."

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

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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 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.

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 $\sim$ 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.

On POA volatility. It seems if the POA is volatile then the assumption that it is not would

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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?

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.

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?

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

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?

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.

P4827, L19 Missing “of” before “HOA”?

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

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

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

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

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

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

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

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

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.

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

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?

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.

P4834, L20 No "d" on "simulate"

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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?

P4836, L3 Replace "predicted reasonably well" with "reasonably good"? I.E don't need "predicted" twice.

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.

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.

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

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.

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

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?

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