**Atmospheric
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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.

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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

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

Major comment 1. The manuscript highlights the ability of the AMS to separate HOA and BBOA. These are also trivial separations for a model study. The authors could therefore make a direct comparison between HOA and simulated POA from fossil fuel sources and BBOA and simulated biomass burning POA. The authors may have already performed such a sensitivity study (by tagging emission sources or turning off individual emissions) (page 4831, line7-8), if not such a sensitivity simulation should be performed. The authors can then compare to HOA and BBOA separately with the source-specific POA simulation on all the platforms and conclude as to which inventories may have weaknesses in which regions / time periods. This also applies to the CO and EC simulation – the percentage contribution of sources at different sites/times may clarify the source of simulation errors.

Response: As suggested by the reviewer, we have added analyses of the simulated anthropogenic and biomass tracers of primary organic matter. We have altered many figures to include a separate comparison of HOA with simulated anthropogenic POA and BBOA with simulated biomass burning BBOA, including Figs. 11 – 16. Figure 12 has also been divided into two figures: one containing sites with PMF available and the other containing sites where only total organic matter measurements are available. Much of the text in Section 5 has been altered to accommodate the new information in these figures.

Major comment 2. There is no discussion of the carbonaceous aerosol simulation. Standard 3D model simulations include hydrophobic and hydrophilic organic aerosols (and EC) which are "aged" with a characteristic e-folding time. Is this the case for WRF-Chem? If so, are the authors limiting their comparison between POA and HOA+BBOA to the hydrophobic component? The aged hydrophilic component may be more directly comparable to OOA in the AMS PMF scheme. If there is no separation in the model, can the authors comment on what fraction of the POA might appear to be HOA+BBOA

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and what fraction might appear to be aged?

Response: Some models treat hydrophobic and hydrophilic carbonaceous aerosols (organic matter and EC) as separate species. This is not the case in MOSAIC, and there is "no separation in the model" as indicated in the reviewer's last sentence. In MOSAIC we assume an internal mixing assumption where all particles within a particular size bin are assumed to have the same chemical composition and the hygroscopic properties of aerosols for a specific size bin are computed as the volume-weighted bulk hygroscopicity for each chemical composition. Since the model employs internal mixing, we cannot differentiate between "fresh" and "aged" organic aerosols. These details have been described in previous publications of MOSAIC, but some additional text has been added to the model description to avoid confusion. We have compared the total POA emitted to HOA+BBOA without attempting to determine what fraction of the aerosol mass is "aged".

Major comment 3. Page 4815: How are the PM2.5 emissions separated in the inventory? Is this breakdown (OC, EC, sulfate, nitrate, ammonium) provided in NEI or was it determined as part of this study? Is the breakdown a constant?

Response: The PM2.5 MCMA emissions inventory was divided into four components: primary organic matter, elemental carbon, other inorganic material, and crustal material. The ratio of the mass emitted by each component to the total PM2.5 mass varied over the Mexico City basin. The Mexican NEI99 emissions; however, did not divide PM2.5 emissions into separate components. Therefore, we computed the average ratio of primary emissions from the MCMA inventory and applied those fractions to the NEI inventory. We also adjusted this ratio somewhat to include a small amount of sulfate and nitrate emissions based on values obtained from urban areas in the U.S. The fraction of PM2.5 apportioned to organic matter, elemental carbon, other inorganics, sulfate, and nitrate was 70%, 15%, and 12.8%, 2%, and 0.2, respectively. Since we did not have any information to vary these ratios spatially, these constant ratios were used throughout Mexico (outside of the Mexico City basin). This information has been

added to the description of the emission inventories in Section 3.3.

Major comment 4. Section 5.1: I agree with the comment posted by Karl that the TOOC discussion is a little unfocused. As the authors do not have a complete organic carbon simulation (neglecting SOA) the overall TOOC comparisons are not very useful. If the authors are attempting to address the potential implications of the carbon budget for SOA formation, it would be useful to see a breakdown of TOOC targeting SOA precursors.

Response: Since the discussion of TOOC is not central to the main points of the paper, we have chosen to eliminate this material. An analysis of the total amount of carbon in the atmosphere is a very useful exercise in evaluating SOA and SOA precursors predicted by models. But given the complexity of VOCs and the number of other sites and aircraft that have available data to compute TOOC, it would be more appropriate to present this material in a separate paper at a later date.

Major comment 5. I'm curious if the authors considered the potential POA source for trash burning? This seems like something that could be quite important in a city like Mexico – was it included in the inventories? Could the authors comment on it?

Response: Our approach was to simply use 'standard' emission inventories developed by others that have been developed. The MCMA 2002 emission inventory document does not indicate that trash burning is one of the sources contributing to PM2.5 or PM10 emissions (it does include it as a source of methane). Of course, there is increasing evidence that this may be a significant source of POA. Smaller combustion, including grass fires, are likely important contributors as well that are currently neglected to our knowledge. We have included a paragraph a the end of Section 3.3 to indicate trash burning as a possible emission sources that are not included in our simulation.

Minor comment 1. Page 4814: Does the MOZART POA simulation match the WRF-Chem simulation? (tracer types and emission inventories)

Response: We use MOZART to simply provide WRF-Chem realistic boundary conditions for POA and other species. One should not expect MOZART POA to match the predictions of WRF-Chem. The grid spacing for the global MOZART model is rather large (hundreds of kilometers); therefore, concentrations over Mexico are likely to be much lower than WRF-Chem simply as a result of averaging over a large area. POA emissions for the MOZART simulation used in this study were also based on a global emission inventory that are not the same as those used in this study.

Minor comment 2. Figure 1d: I recommend adding text to the caption to clarify that the fire hotspots were obtained by MODIS

Response: Phrase has been included to clarify MODIS source of biomass burning locations.

Minor comment 3. Including both Figures 4 and 5 seems a bit excessive. I'd recommend removing Figure 4 and abbreviating the discussion (as this is a comparison with assimilated winds).

Response: We wish to retain both figures, since one figure illustrates model performance for surface winds and the other figure illustrates performance of the simulated winds aloft. Uncertainties in simulated winds are often larger close to the ground, especially in areas of complex terrain. Therefore, it is important to understand both in relation to the transport of trace gases and aerosols.

Minor comment 4. The correlation coefficient and bias numbers should be added to Figure 5.

Response: Added correlation coefficient and bias to the figure. A sentence has also been added to Section 4.1 regarding the statistics.

Minor comment 5. Figures 9 and 16 are difficult to read. I'd recommend either replacing all the bars with the mean comparisons for each platform or greatly expanding the vertical axis to make the quantities more legible.

Response: The vertical extent of Figure 9 has been expanded and the text is somewhat larger. Figure 16 has been changed significantly to address other comments, and these changes also made it more legible.

Minor comment 6. Page 4826, 1st line: I believe there's a typo and the text should indicate a negative bias (an underestimate by the model @ Paso). Also, comparisons at T2 are not included in Figure 7. Please add to the figure or modify the text.

Response: Yes, that was a typo and the text has been corrected. CO was not measured at T2 (but EC was measured) so that phrase was removed.

Minor comment 7. I recommend adding the correlation coefficients between the model and HOA+BBOA for the entire timeseries (and the individual HOA and BBOA with the fractions of the relevant simulation after the sensitivity simulations are completed, see point #1 under major comments) to the top of Figure 13.

Response: This figure as been revised to include individual HOA and BBOA components, and the biases and correlation coefficients have been included as well. Some text has also been added regarding the correlation.

Minor comment 8. Page 4835, line 6: SVOC and IVOC are not included in the model, but nor are they measured. This should be clarified.

Response: Section 5 has been removed to address major comment 4, so this text no longer appears in the paper.

Minor comment 9. Page 4838: It would be useful to indicate the size range of POA used for model comparisons far before the Discussion text. I was wondering about this in the model description section.

Response: The model description section states the number of size bins employed between the cut-offs of 0.39 and 10 micrometers. Rather than add a description of the cut-offs in that section, we added a few sentences near the beginning of Section 4.4 to clarify how the model was compared to the data.

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