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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modeling the impacts of biomass burning on air quality in and around Mexico City" *by* W. Lei et al.

Anonymous Referee #1

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Review of "Modeling the impacts of biomass burning on air quality in and around Mexico City" by W. Lei, G. Li, and L. Molina

Calculations using WRF-CHEM are used to determine the perturbations to ozone, POA, SOA, TOA, and BC due to open fires, trash burning, and, in less detail, biofuel use over different spatial domains and time periods. Of particular importance are the ground sites with AMS measurements; T0 having data that covered both simulated episodes, and T1 having data for one episode. The general conclusions are that fires are important, more so for open burning than trash burning and more so for POA than SOA. The fraction of organic aerosol contributed by fires is significantly higher than the fraction contribution to EC.

As the authors acknowledge, results are highly dependent on emission rates. A major

concern that I have with the calculations is the justification for an alternative diurnal profile for emissions. The alternative profile is meant to provide a better description of nighttime emissions from smoldering fires. Fire counts are from Modis, which is daytime only. However, the default diurnal cycle is from GOES. Is the implication that the default diurnal cycle is in general, incorrect. Or are nighttime smoldering fires being put into the model to simulate features particular to this time and place where there is observational evidence for such fires. Going one step further, is the observational evidence from visual reports, measured trace gas excursions, or aerosol observations. Fig. 6, shows that the adjusted emission profile makes a large difference on a few days, generally to bring observations and calculations into better agreement. It is difficult to see from the Figure what the integrated effect is.

If the effect of overnight smoldering emissions is due to high concentrations trapped under a low boundary layer, there should be evidence in terms of diurnal cycles. A comparison of simulated and observed diurnal cycles should be provided.

A gauge of whether the emission time dependence is reasonable is part of a larger concern that I have. This is a complicated calculation. Results seem reasonable. But what have I learned, that is not contained in previous studies? Certainly different regions and categories of burning are being considered here. What about processes? Has a convincing argument been made concerning the importance of overnight smoldering emissions? What is the SOA/POA ratio in biomass burning aerosol? This result can be obtained from percent changes in POA, SOA, and TOA (mentioned below). Does the above ratio vary with location? How does this ratio depend on the models chemical mechanism and VOCs that are co-emitted in fires? Are the ratios calculated at varying locations a strong function of plume age?

I believe that additional material on some of the "why" questions (which don't have to be the ones mentioned above) are needed in this manuscript.

General questions

Model results are generally presented as the contribution of a source category (such as biomass burning = open flames + trash burning) to POA, SOA, TOA, and EC, for different time periods and locations. The percent changes in POA, SOA, and TOA combined with the definition of TOA (=POA+SOA) are readily manipulated to give the ratios POA/TOA and SOA/TOA before and after biomass burning emissions have been added to a base case simulation. The relative proportions of POA and SOA are important because they give the reader a sense of the amount of photochemical processing and they provide an additional number that can be compared with observations as seen through the lens of PMF analysis. The paper, as written contains many percent contributions, to the point of confusion for me (but perhaps not for other readers), so I am suggesting that POA/TOA and SOA/TOA be presented only for a subset of the data.

Are ug/m3 at ambient T and P or at STP?

Ordered list of questions and comments.

Abstract: It is difficult to follow the presentation of percent contributions. I am not sure if MCMA and MCMA plus surrounding areas are being used interchangeably. Rather than cite a string of 4 components and 4 percentages and tie them together with "respectively", I recommend something like this: open fires plus trash burning increase POA by 60%, SOA by 22%, TOA by 33%, and EC by 22%.

The summary section also has multiple lists of percentages in four categories. I recommend a summary Table, without repeating all of the numbers in the text.

It is not obvious that the statement on line 15-16 that SOA formation increased OA by about 10% from open fires and 5% from trash burning is consistent with the values given above. (I did the math, and they are). I would tie these statements together by e.g. Of the 22% increase in SOA (equivalent to a 15% increase in TOA) predicted for biomass and trash burning about 2/3 is from biomass and 1/3 from trash.

page 22896 lines 21-23. Abbreviation TB This is perhaps, obvious. The first sentence uses the term trash burning and the second sentence uses the abbreviation TB. The first sentence should read "trash burning (TB)"

page 22896 line 22-23. impact of TB on PM2.5 and chloride will be addressed in a companion paper. In the abstract and immediately previous to this sentence you state that this paper covers TB. For clarity, I recommend modifying line 22-23, e.g. Additional analysis of TB impacts on PM2.5 and chloride are presented in a companion paper.

page 22896, line 26, regarding biofuel. My personal preference would be to replace "we will attempt to provide a first-order" by "we only attempt a first-order"

Page 22899 line 17 – 24. Do, I have this correct? The model contains material not in "conventional calculations" equal to 3.5 times POA not corrected for dilution (additional SVOCs = 2 times conventional POA and co-emitted IVOCs = 1.5 times conventional POA)? The total amount of material (POA+SVOC+IVOC) is 7.5 times POA emissions not corrected for dilution.

What is the relation between POA emissions in your calculations and what I am calling "conventional" POA? Some part of the difference between 7.5 and 3.5 is emitted POA in your model. What is the other part? Would that other part also be in a model that did not take into account additional low volatility VOCs? These emissions make a major difference in SOA predictions and it is important for the reader to qualitatively know what they are, without reading cited papers.

page 22900 line 13 – 15 regarding alternate emission profile Would it be just as likely for GOES to miss a smoldering fire during the daytime as in the nighttime?

Page 22901 Section 2.2.3 Trash burning First paragraph is about trash burning. Later paragraphs fold in material on other sources.

page 22902 line 18. 35% uncertainty Unclear whether this refers to PMF method alone or includes accuracy of AMS measurement. My opinion is that 35% is about right for

the uncertainty of the measurement. Others may be more optimistic. A reference would shift the blame.

page 22906 line 16 – 18 Lack of correlation between T0 and T1. Image plots show a time lag between polluted air hitting T0 and T1.

page 22906 line 18 “due to that the” needs rewording

Page 22907 line 12 “No comparisons at T1 are presented in this study ... limited availability of AMS OA measurements” There is one episode at T1, one third of the AMS data set. This is a lot of data to throw away. Some of it is shown for other purposes.

Page 22907 line 19 – 24 OH recycling, semi-volatile VOCs did not consume OH I don’t understand. OH + VOCs don’t usually consume OH, but instead are part of a chain reaction. How does the chemistry of intermediate and semi-volatile VOCs differ from other mechanisms.

Page 22908 line 26 – page 22909 line 2 I don’t understand how model underestimation of EC at T1 probably contributes to this result.

Page 22910 line 8 typo replace conductive with convective

Page 22910 line 27 typo replace Table 3 with Table 5a

Page 22912 line 12 TB has smaller influence on regional scale Where there TB emission in calculation outside of MCMA. In MCMA they were proportional to population and a few other variables

Page 22913 line 2 “PEC emissions” New abbreviation

Page 22915, line 25 typo emissions to another, should be emissions as another

Page 22917 lines 23-27. There is general agreement between model and observed BBSOA at T0 and T1. Does biofuel SOA get classified with PMF along with SOA from

open flames and TB? If so, does this imply that biofuel SOA is small, or that it is within uncertainty of calculation?

Table 1 What are ranges for % contribution?

Table 4 For those looking for red and green traces in ozone figures, it would be useful to mention that they are so close that red hides green.

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