

Interactive comment on “Global combustion sources of organic aerosols: Model comparison with 84 AMS factor analysis data sets” by A. P. Tsimpidi et al.

Anonymous Referee #2

Received and published: 26 March 2016

This paper presents an analysis of a global chemical transport model's predictions of organic aerosol, through comparison with data from many AMS across the globe. Although it is encouraging to see these AMS data being used, I found the analysis in this paper disappointing, and in the end I cannot see that we learn very much. I cannot recommend this paper for publication for the following main reasons:

1. Much of the focus is on differences between urban, urban-downwind and rural OA components, but the horizontal resolution of the model used, 1.875×1.875 degrees (ca. 200 km) is wholly inappropriate for analysis down to the urban scale. Even Mexico city has dimensions of 10s of km, not 100s km.

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The authors admit on p18:

'.. the model cannot reproduce the concentrations of POA and SOA over urban locations due to its coarse resolution'.

They therefore chose not to use urban results when looking at seasonal patterns. However, if the urban results are not valid for seasonal comparisons they are clearly not valid for annual comparisons either!

(I also wonder why such a low resolution was used. This model has previously been run at higher resolutions. Presumably CPU time was not a big issue, since we are looking at the results of just one model run. For the results of sensitivity tests on IVOC/POA ratios and deposition parameterization we are referred to papers which are yet to be written.)

2. Essentially the whole paper builds upon global averages of OA results, and we are given no idea of how the model performs in different parts of the world. Global averaging can mask a whole slew of incorrect regional results, and with a pollutant as complex as OA this is a dangerous procedure. As can be seen in the (originally not-mentioned!) papers (Karydis et al., Pozzer et al.) where some model evaluation can be found for the inorganic components of this model, the model displays strong under and over-predictions of species such as sulfate and nitrate in different parts of the globe.

I comment more below on the author's inattention to model evaluation (and their response to my original criticism of this), but although Tables S1-S3 now give the site by site comparisons, it is a daunting job for the reader to work out the spatial patterns in model under or overestimation. How does the model perform in North America, Europe and Asia? I would have liked to see global bias maps (of at least total OA, ideally components) compared for the different types of stations. Ideally

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with separate presentation for summer and winter months, since the biomass burning signal is so large.

Given that different seasons typically have very different sources of SOA, I would have liked to see maps (or mapped pie-charts or similar, as per Jimenez et al., 2009) of model bias and correlation for these different seasons.

3. I was also surprised to see no mention of, let alone comparison with, ^{14}C data. For example, Hodzic et al. (2010) used published results from measurements to provide a comprehensive view on the fraction of fossil and biogenic carbon across the globe, and in many cases for both summer and winter periods. Such data would have provided a very good check on some of the model predictions presented here.
4. I was not convinced by several aspects of the author's reply to my query concerning evaluation. I give here the authors comments in italic, and my replies in normal font:

In our recent paper (Karydis et al., 2016) we have found that EMAC tends to overestimate the inorganic aerosol components over Europe while over North America and Eastern Asia, the inorganic aerosol concentrations are slightly underestimated by the model.

Well, Karydis suggests a factor 3 over-prediction of nitrate and sulfate in Europe, and a factor 2-3 under-prediction in East Asia, which is more than 'slightly' underestimated. Although such discrepancies are probably not surprising for global models, their magnitude raised serious questions about the interpretation of much more complex OA comparisons.

Given that the performance of the model is increasingly realistic with increasing resolution, the acceptable model performance shown from all the previous model evaluations (in both higher and lower spatial resolutions than the one used in

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our work) should be enough to verify that EMAC can cope with the inorganic components under any resolution.

I completely disagree with this sentence. As noted above, one model setup can produce factor of three over-predictions and factor two under-predictions at the same time. It is quite likely that changing resolution might help some problem areas but exacerbate others. If for example there are problems with vertical dispersion in the model (quite likely with any model), then a change in horizontal resolution can easily cause incorrect responses.

Increasing resolution can also expose other problems which lower resolutions might mask out, for example if the emissions are incorrectly distributed.

Furthermore, there is no chemical or thermodynamic link between the inorganic (GMXe) and organic (ORACLE) aerosol modules used in this study. Therefore, we strongly believe that a comprehensive evaluation of inorganic components in our manuscript will disorientate the reader without providing him/her any useful information regarding the behavior of the OA in our model system.

Well, there are clear links between the inorganic and organic pollutants, which include meteorology (including dispersion issues, mixing heights, etc.), emissions, and atmospheric chemistry (OH, O₃, NO₃). If EMAC is under-predicting e.g. NO₂ in an urban area, I would expect it to under-predict OA too, especially near-source POA levels.

5. The discussions in e.g. 5.2 explain any discrepancies solely in terms of SOA mechanisms or rather particular emissions problems (e.g. cold start). There are many reasons why modeled and measured OA components don't match, including for example dispersion or deposition processes, which is why I don't think we learn very much from rather broad-brush OA comparisons alone.
6. This paper focuses on the primary emissions, with presumably other papers to come addressing biogenic and suchlike. One of the biggest assumptions with

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POA is the factor 1.5 used to get IVOC emissions. This factor is very uncertain, and I would have expected a paper concerned with POA to address this uncertainty directly. Instead, we read on p.10 (L277-278) that this issue will be addressed in another paper which is in preparation. This is surprising, and again makes me wonder what this paper is actually showing, apart from largely unverified model predictions.

7. There is always the danger with SOA modeling studies that a lot of colorful maps are presented, and various numbers given, but the reader never knows if these results are meaningful. We have seen global modeling papers for OA for about two decades now doing this, often with claims that 'this time' the results are reasonable. With time such results change markedly though as new understanding becomes available, often with dramatic results. For example, the recent paper by Hodzic et al. (ACPD, 2015) is suggesting that SOA processing and losses should be much faster than previously assumed.

With AMS data one has the possibility to try to pin down model performance in a much more robust way that was previously done with filter measurements. By ignoring the spatial and temporal variations seen in these AMS data the current paper missed a real chance to show how well their model performed, and thus how seriously we should take the results.

Other points:

- General: The paper makes use a large fraction of older references. Although I like to see citation of original or ground-breaking papers, I also expect to see that the authors know about and have incorporated recent advances in their work. The cited references throughout the paper need to be checked and updated where possible. (I give some examples below, but in general I worry when literature is more than 10 years old in this field.)

- Although the focus of this paper is on anthropogenic SOA, the treatment of biogenic VOC is rather confusing. On p9 for example we read that the production of RO₂ after oxidation of SVOC and IVOC has been omitted because the OH-sink is so low. What about forested regions though, where there are plenty of papers showing that the OH sink from BVOC can be very significant indeed (Kurpius & Goldstein, 2003, Mogensen et al., 2011)? The Tsimpidi et al. (2014) paper which introduced ORACLE has reaction R3 which also seem to ignore RO₂.
- p2, L42-43: Technically, aerosol is a mixture of gas and particle, and some authors use the term POA to include both phases, so it is good if the authors state clearly at the beginning that for them POA means the particulate fraction of primary OA.
- p3, L46. Please be explicit. You mean particulate OC, not gaseous VOC species.
- p3, L46-50. Again, old literature (>10 years) is used. These numbers need to be updated, e.g. with those from Tsigaridis et al. 2014.
- p3, L50. What is the reference for the 13-57 Tg/yr?
- p3, L53: May et al., 2014, and/or Ranjan et al. (2012) could also be cited here for a more recent evaluation.
- p3, L564 Again, only old references are used (>14 years!). There has been a lot of work and progress (even 'rethinking') since these Schauer evaluations.
- p4, L109. It is not true that OOA has the same diurnal pattern as other secondary organic PM components. Indeed, not even sulfate and nitrate have the same pattern as each other. The authors even mention later in the manuscript that one can usually observe different diurnal variations for SV-OOA and LV-OOA.

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- p5, L120-122. These statements need to be updated, e.g. Tsigaridis et al. 2014 show that this commonly repeated statement that CTMs underestimate OA is not always true.
- p5, L131-132. Donahue didn't 'assume' that emissions were distributed in bins. The bins are a modeling approach applied to a what he knew was a continuous distribution of volatilities.
- p5, L141 on. Again, the Tsigaridis paper is the current best source of information on global SOA models.
- p6, Sect. 2.1. As mentioned in my earlier ACPD comment, I would have expected a model description to point readers towards works that could prove the validity (or at least document performance) of any model. This manuscript is remarkably free of the kind of information on model performance (also for inorganic components) that I expect to see in any model study.
- p7 - L204. The sentence about LVOC not being allowed to participate in photochemical reactions begs the questions of how SVOC or indeed any semi-volatile compounds are allowed to participate, and how. Do SVOC and IVOC deplete oxidant concentrations, and if so, what happens to products and e.g. RO2 formation?
- p8, L211-216. The authors claim that a reduction in volatility by a factor of 100 is common in global models, but cite just their own code and one other paper (Pye and Seinfeld, 2010). Can the authors justify the word 'common'? In fact, this reduction of a factor of 100 for SVOCs and IVOCs upon oxidation is rather extreme. Although I know this has been dealt with to some extent in the 2014 paper, another sentence or so of explanation would help the reader of this paper.
- p8, L215. The statement about regional models and the following sentence miss an important point. The regional models have usually followed the suggestions

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of the groups that developed the VBS ideas (Donahue, Robinson, etc.), and who worked directly with smog-chamber data. As noted in Donahue et al. (2009), the phase partitioning of a complex mixture can be accurately described by lumping material into volatility bins separated by an order of magnitude (at 298 K). This is not only convenient; it is a nearly optimal distribution. The cruder methodology (bins spaced by two orders of magnitude in C^*) being suggested in the ORACLE scheme may well be sufficient for global models given the large uncertainties of any SOA scheme, but is not optimal. Indeed, a lot of atmospheric 'action' may be expected to take place over quite a narrow range of concentrations between say 1 and 10 $\mu\text{g}/\text{m}^3$, a range which isn't resolved by the factor 100 spacing of ORACLE.

- p13, L381. This statement, that the fOA/OA fraction is 0-10% over boreal regions is incorrect. Values seem to be 0.3-0.5 over much of the Eurasian boreal forest. Which is rather surprising, isn't it?
- p17, L517. The Denier van der Gon 2014 ACPD paper made it to ACP in 2015.
- p23. Why would aqueous-phase and heterogeneous oxidation be important in winter but not summer? Provide some citation to back up such statements. I would also suspect that the factor 100 spacing between volatility bins would have some implications for summer/winter differences in model performance.
- p34, Table 1. Add BSOA also.
- p35, Table 2. The caption mentions 84 stations, but the table lists 82 in the upper rows and 60 in the lower. Explain.
- Figs: The quality of the Figures should be improved. In Fig. 4 the color scale leaves almost the whole map as different hues of blue. Also, although acroread shows Figs. 1 and 2 fine, my print-out has black-backgrounds and is almost unreadable. I suggest a change of Figure format for this.

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