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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol mass spectrometer constraint on the global secondary organic aerosol budget" *by* D. V. Spracklen et al.

Anonymous Referee #2

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The manuscript describes a modeling study that aimed to reduce the uncertainty on the secondary organic aerosol (SOA) sources, both in magnitude and origin. The method is based on a multi-simulation approach, where several arbitrary selected cases were tested, and then were optimized, to match measurements. A different set of measurements was used to test the optimization results. The concept of this work is very good; it appears to be a very clever way to constraint SOA production. Nevertheless, there are some major issues that need to be addressed before this paper is accepted to ACP.

General comments

1) The concept of this work is very good; the approach though seems not to be the proper one. The questions the authors tried to answer belong to an inverse modeling study, not a traditional one. Having measurements and trying to constraint sources is

exactly what the inverse models are trying to constrain. Using a forward modeling technique like the one described in the manuscript, one trusts that the spatial distribution of sources is correct, and only the magnitude is the free parameter. This applies mainly to the primary sources of organic aerosols, but also at the precursor volatile organic compounds of SOA.

2) A potentially significant source of SOA, that of IVOC, is not taken into account. In addition, the semi-volatile nature of primary OA is treated as a sensitivity study, and not as the default one. These two can significantly alter the OA spatial and temporal distribution, drastically affecting the results of this study. Especially concerning volatility, there are some very important free parameters in their calculation that need to be constrained, like the volatility distribution of the emissions, the temperature dependence of the volatility and their volatility change with time. Trying to estimate the sources of SOA using the traditional approaches appears to be problematic.

3) It is not very clear where the anthropogenically controlled SOA come from. The authors mention that they are linked to anthropogenic emissions, but only 10% is urban. Where does the carbon come from? If only from anthropogenic sources, then the 100Tg/a of SOA seem excessive, even with an OM/OC ratio of 2. This would mean that almost all primary anthropogenic emissions have a close to 100% SOA yield. On the other hand, if the carbon comes from biogenic sources that mix with anthropogenic ozone, NOx and other pollutants, then this source double-counts the biogenic carbon fluxes, since they are also being considered as monoterpenes and isoprene. Scaling the IPCC CO emissions from which sources gave the VOCa emissions distribution? How can it be verified that scaling CO in one hand and using aromatic emissions in the other hand does not double-count carbon sources?

4) The whole study is very strongly related with the host model. Such fine tuning of sources depends on the underlying climate, transport, chemistry, removal. A critical question which was not addressed is how much this optimization depends on the model. Understanding that it would be unrealistic to repeat this experiment with an-

other model, one should try to implement a few key simulations from table 2 to a very different global model, and compare the results with the present study. On top of that, it should be extremely clear in the manuscript (abstract, body, conclusions) that this optimization is not meant to be a generic suggestion; rather, it is a very model specific calculation of some, but not all, uncertainties in global SOA sources. If a second model is included in the study, then, depending on the results, one might extract a more general conclusion. At the present state of the manuscript, the authors should discourage the reader from using their numbers as a suggested range of SOA sources.

5) The SOA production chosen, as shown in Table 1, does not agree with literature data. It had been shown by numerous publications the last 15 years that using a single product to simulate SOA formation from a given precursor VOC is insufficient.

6) Given the large uncertainties of the study, I suggest to remove section 3.7. The SOA sources are so heavily tuned and fit to the GLOMAP model, that calculating a radiative forcing out of it appears premature.

Specific comments

1) Pye and Seinfeld (2010) are not the only ones that made a global modeling study on IVOC and the semi-volatile organics, Jathar et al. (ACPD, 2011) have also done one.

2) Page 5704, lines 8-10: Do you have such a process in the model?

3) Page 5704, lines 15-18: Is this scaling justified for non-polluted regions? How can a correlation of CO and aerosols be justified, given their very different lifetimes?

4) Page 5704, lines 18-22: The authors seem not to trust the EDGAR global numbers, but do use their spatial distributions. How can this be justified?

5) Page 5705, first paragraph: Volatility is extremely important. Pye and Seinfeld (2010) and Jathar et al. (2011) get different answers by using the volatility change with aging differently. Tsigaridis and Kanakidou (2003) get different vertical distributions by changing just the temperature dependence of volatility, and about a factor of 3 in-

crease in SOA production by not allowing them to evaporate. In a global modeling study, aerosols might have plenty of time to evaporate and recondense. Please comment on the uncertainty introduced by assuming no evaporation.

6) Table 2 is very hard to read. It belongs to an appendix, and it has to be replaced by a more condensed and elegant presentation of the simulations performed and their results. In addition, the criteria used in the selection of simulations have to be mentioned. All possible combinations of cases have been simulated? If no, which were the criteria used for selecting which ones to simulate? How was it tested that no bias was introduced by not simulating every possible combination?

7) Page 5706, line 5: How many Tg/a of SOA from monoterpenes?

8) Page 5707, lines 1-4: Several papers presenting AMS results show a third component, in addition to HOA and OOA, that of BBOA, from biomass burning. Is it valid, based on measurements, to assume that 100% of BBOA is HOA?

9) Page 5707, lines 13-15: How about Heald et al. 2005 that shows a very large discrepancy between measurements and models in the free tropospheric OA?

10) Page 5708, lines 11-13: The model appears to be as “successful” with HOA as it is with SO4.

11) Page 5709, lines 17-23: The tropical AMS measurements (OP3 and AMAZE) were during the wet season, thus no biomass burning. Which other measurements were used to constrain the biomass burning source in the tropics? If none, then the global biomass burning source was constrained from mid-latitude and boreal fires, which is not correct.

12) Page 5709, line 27: “heterogeneous oxidation”: why not homogeneous oxidation as well?

13) Page 5713, line 29 to page 5714, line1: I disagree with this statement. Most models have the same aging parameterization, which is that of Cooke et al.: Globally constant

lifetime against aging of about a day. The real uncertainty should be much larger.

14) Page 5714, line 3: The sentence should start with “We arbitrary estimated”. If the choice was not arbitrary, it should be justified.

15) Page 5715, line 8: “To remove” should be “To correct”. The bias cannot be eliminated, since lifetime affects the distribution.

16) Section 3.4: Is the optimization affecting the lifetime of OA? If yes, how much? If no, why?

17) Page 5716, lines 15-16: For SOAb, there is a factor of 5 difference. It might not be so important globally, but it clearly is regionally.

18) Page 5717, last paragraph: Not all authors use the same definitions and precursors for SOA. Please confirm that the comparison is valid and mention it in the text.

19) Page 5719, 2nd paragraph: How do the 14C measurements correlate with IMPROVE and AMS? Is there any overlap between them?

20) Page 5720, lines 17-19: The 10% urban SOA appears to be arbitrary. See also general comment #3.

21) Page 5721, line 2: “substantially larger enhancement”: how much?

22) Table 1, reactions 9 and 10: Are these the ones that are used to simulate the anthropogenically influenced SOA? Why regenerating VOCa and SO2? Why there is no oxidant dependence? How important these reactions are to the global SOA? More explanations are needed here.

23) Figure 8a,d: There is a peak above Mexico. Is this due to the Mexico City very high measurements? Maybe it biases the whole optimization high?

24) Figure 9d: Aged POA can be seen downwind of biomass burning regions, but not downwind of anthropogenically influenced regions. Is this a color scaling issue?

Technical corrections

1) Section 3.3 should be before 3.2, there is no point optimizing the non-representative results.

2) Page 5713, lines 8 and 19: “OOA” should be “SOA”.

3) Page 5714, line 15: “may introduce” should be “introduces”.

4) Page 5717, line 7: “United States” should be “western US in summer”.

5) Table 2 legend: “ SOA yields. . .percentage”: I do not understand this sentence.

6) Figure 6: The concentration bins are extremely large.

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