

Interactive comment on "Influence of biomass burning vapor wall loss correction on modeling organic aerosols in Europe by CAMx v6.50" by Jianhui Jiang et al.

Anonymous Referee #1

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Jiang et al. have performed a modeling study, first to determine parameters to model secondary organic aerosol (SOA) formation from biomass burning and second to use those parameters to study impacts on ambient organic aerosol in Western Europe. They find that accounting for vapor wall losses in chamber experiments seems to generally improve model performance for the OA system (OA, primary OA, and SOA) over the modeling domain, although there are exceptions to the improvement.

This study attempts to better understand the contribution of residential wood combustion to OA concentrations in Western Europe. RWC, referred to as biomass burning in this study (if this is true, this needs to be explicitly stated in the paper to avoid this

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source being mistaken for wildfires), is a major source of air pollution in this part of the world and hence the study is well motivated. The chamber modeling and atmospheric modeling approach is justified although a lot of details are missing. The interpretation largely follows from the model results but could benefit from a more detailed description. I am inclined to recommend publication to Geoscientific Model Development after the authors have had a chance to respond to my comments.

Major comments:

1. SOA precursors: I am familiar with this group's earlier work (i.e., Bruns, Stefenelli) where the SOA formation was modeled by knowing the detailed gas-phase precursors and their corresponding SOA mass yields. However, in this work, it is not at all clear what the SOA precursors are and how they are dealt with, with respect to their emissions, oxidation chemistry, and SOA parameters. For instance, what are the SOA precursors? How are their emissions determined? What are the oxidation rate constants? Is there any accounting done for precursor composition? Are all SOA precursors lumped into a single model precursor? Or are all species modeled separately with their own oxidation rate but with the same volatility distribution for their oxidation products? Were different functional forms of the volatility distribution explored and what was the rationale to pick a normal distribution? Answers to these questions and more need to be described in much more detail in the methods section for the reader to follow the general approach. A further point of confusion is how this is then translated to be used in CAMx in Section 3.2

2. OA schemes: I was a little confused about the diversity of schemes tested in this work, particularly if the goal was to examine the specific influence of vapor wall losses on ambient OA concentrations. In other words, shouldn't the work have focused only on VBS_WLS and VBS_noWLS? At present the other simulation results are preventing a clear interpretation of the model results. Furthermore, I was concerned about the treatment of SOA precursors in the two VBS parameterizations: VBS_BASE and VBS_3POA. There is emerging evidence, including that from previous publications

from this group, that SOA precursors from biomass burning are poorly represented in atmospheric models. Along with that evidence though, are detailed observations of what those SOA precursors are, as SVOCs, IVOCs, and VOCs; see for example Hatch et al. (ACP, 2015), Hatch et al. (ACP, 2017), Jen et al. (2019), Koss et al. (ACP, 2018), Sekimoto et al., (ACP, 2018). These studies provide sufficient data to inform the treatment of SVOC, IVOC, and VOC emissions in this work and hence I am concerned that the treatment in this work is a little dated. The current approach to determine IVOC emissions by multiplying POA by 4.5 and SVOC emissions by multiplying POA by 3 seems a bit too much. These S/IVOC estimates need to be reconciled with the extensive literature that I have alluded to earlier. Additionally, I am unclear how the VOCs are dealt with in CAMx. Are these mapped to the traditional SOA precursor species (e.g., benzene, toluene, etc.)? Finally, are vapor wall loss effects accounted for, for the non-biomass burning sources and SOA precursors? What approach is used to do this?

Minor comments:

1. Lines 37-41: Citations are a little dated, replace with more recent studies/reviews.

2. Line 52: Akherati et al. (ES&T, 2020) recently looked at the impacts of vapor wall losses on SOA production from biomass burning emissions. Consider citing.

3. Line 53: Zhang et al. (PNAS, 2014)'s conclusions are sensitive to NOx where the enhancement is much lower at high NOx conditions. Consider stating this.

4. Line 59-60: Not sure I agree with this statement of an enhancement larger than 4.

5. Line 76: Remove 'datasets from'.

6. Line 86: Unclear what 'be diluted' means. The experiments need to be described in more detail in Section 2.1 for the reader to connect the modeling approach to the how the experiments were conducted.

7. Lines 87-92: What is the rationale for not accounting for losses of condensable

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vapors to the particles on the walls?

8. Line 97: Both Cappa et al. (ACPD, 2020) and Akherati et al. (ES&T, 2020), in chamber experiments performed on biomass burning emissions, found little evidence for POA evaporation with dilution suggesting that the POA was either composed of low-volatility material or there were limitations to evaporation linked to the phase state. What are the implications of the findings from these two studies here?

9. Line 100: It would be useful to compare the vapor wall loss rates used in this work (derived from Bertrand) to similar estimates now made for other chambers: Zhang et al. (PNAS, 2014), Krechmer et al. (ES&T, 2016), Nah et al. (ACP, 2016), He et al. (ESPI, 2020), and Akherati et al. (ES&T, 2020). There might also be a few additional studies from Carnegie Mellon that you could look into. How does that comparison look?

10. Equation 2: Why is particle wall loss not included in here? If the measurement data are already corrected for particle losses, what estimates were used, i.e., w=0, w=1, or average?

11. Line 118: Krechmer et al. (ES&T, 2016) have argued that C_wall needs to be modeled as a function of C^{*} based on observations of loss rates of oxidation products with different C^{*}. Can this also be modeled in this work?

12. Section 3.2: Perhaps, focus this section on the VBS_noWLS and VBS_WLS and frame the other schemes as sensitivity or legacy simulations?

13. Line 162: By how much was the aerosol yield scaled by to account for vapor wall loss?

14. VBS_WLS: How does one think about the relatively higher NOx conditions in the chamber experiments and the much lower NOx conditions in the atmosphere while translating the SOA parameters from the chamber to the atmosphere? How can this influence be further studied via sensitivity simulations?

15. Lines 202-203: What was the OM loading in the other experiments that is com-

pared to here?

16. Line 215: Is the temperature effect on gas-phase chemistry taken into account?

17. Section 4.1, last paragraph: I think I understand what you mean but try to explain the 'vapor wall loss yield' results better since one would expect the yields to be lower when accounting for vapor loss to the walls.

18. Figure 3(a): Are those emission factors or production factors for SOA?

19. Figure 5: What is the R²? Can you describe the model skill and how it compares across schemes, cities, and other comparable models?

20. Figure 6: It is surprising to see so much SOA production compared to POA even in the winter. Are aqueous pathways for SOA production accounted for? Were the chamber experiments done under 'dry' or 'wet' conditions?

21. Figure 9: Is this just for winter or all year round?

22. Line 283: Can you make emissions maps to make this point about the dominance of biomass burning emissions in certain locations clear?

23. Section 4: The paper would benefit from doing a sensitivity simulation where biomass burning is eliminated from the emissions to understand the relative contribution of this source to total OA/POA/SOA.

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