Anonymous Referee #2

In this paper, Tsimpidi et al. performed different sensitivity tests with the global chemistry-climate model EMAC in order to investigate the main parameters affecting the evolution of organic aerosol from combustion sources. Different assumptions on primary organic aerosol emission inventories, volatility distributions and reaction rate constants of SVOCs and IVOCs against OH are investigated. In addition, the authors deployed alternative aging schemes as well as different values of the Henry's law constant to test the effect of wet removal of SVOCs and IVOCs from the atmosphere. The ORACLE module, based on the VBS framework, is used within EMAC to model the evolution of OA in the atmosphere and results from the sensitivity tests compared against a comprehensive set of AMS measurements performed during 2001-2010.
The paper deserves publication, the results are well presented and the adopted schemes are appropriate for the analysis.
I recommend the paper for publication after considering the minor comments below.

We would like to thank the reviewer for his/her positive response. Below is our point by point response to the reviewer's comments.

1. *Line 33: a more recent reference is needed.*

   The sentence has been changed to "Organic aerosol (OA) is an important constituent of the atmosphere, contributing about 50% of the total submicron dry aerosol mass (Zhang et al., 2011) with major impacts on human health and climate (IPCC, 2013; Lelieveld et al., 2015)."

2. *Line 38: "which can reduce their volatility". In recognizing that the main point of the sentence is to describe the formation of SOA, it would be desirable to mention the increase in volatility due to fragmentation as well.*

   Following the suggestion of the reviewer we have rewritten the sentence as follows: "The co-emitted organic vapors can undergo one or more chemical transformations, which can alter their volatility due to functionalization (reducing their volatility) or fragmentation (increasing their volatility). The oxidation products with lower volatility can be transferred to the particulate phase forming secondary organic aerosol (SOA)."

3. *Line 48: Please consider adding Jo et al., 2013 who has also investigated the effects of chemical aging on global secondary organic aerosol using the GEOS-Chem model and compared the model results against AMS datasets.*

   Done.

4. *Line 142: What is the thickness of the first layer? Please add this information.*

   It is 68 m. We have added this information in the revised manuscript.

5. *Line 163-166: "The volatilities of SVOCs and IVOCs are reduced by a factor of 10^2 as a result of the OH reaction with a rate constant of $2x10^{-11}$ cm^3 $molecule^{-1}$ s^{-1} and a 15% increase in mass to account for two added oxygens (Tsimpidi et al., 2014)". Does the model include any fragmentation pathways as well? Please specify if fragmentation is directly/indirectly accounted for.*

   The model does not include explicitly the fragmentation pathway. This has been indirectly taken into account by assuming that the functionalization and fragmentation processes result in a net average decrease of volatility for SOA produced by SVOC/IVOC and anthropogenic VOC and no

net average change of volatility for SOA produced by biogenic VOC (Murphy et al., 2012). This information has been added in the revised manuscript.

6. *Line 170: Were shipping emissions taken into account?*

   Yes. Shipping emissions are part of the CMIP5 RCP4.5 emission inventory.

7. *Line 359-362: "On the other hand, OOA concentrations are underpredicted (-31%; Table 3) indicating that the model may be missing an important source or formation pathway of SOA especially in winter (Tsimpidi et al., 2016) or may be removing the corresponding pollutants faster". Please add the uncertainties in SOA yields due to wall loss in chambers as another possible reason for the underprediction of SOA. In the authors opinion, how much do vapor wall losses influence their results?*

   Thank you for the helpful suggestion. The loss of semi-volatile vapors to the walls of laboratory chambers has been added as a possible reason for the underprediction of OOA. According to Zhang et al. (2014), these vapor losses can lead to substantially underestimated SOA formation.

8. *Figures 4-5-6 and 7: In general, it seems that for all the sensitivity tests almost no changes are observed in the Scandinavian region. Is this simply because of the low SOA concentration predicted in this area of the domain? Or are there other reasons?*

   Figures 4-7 depict absolute changes of OA concentrations for each sensitivity test; therefore, changes are low due to the low OA concentrations predicted by the model over the Scandinavian region (Figure 3) in the base case scenario. Changes are only noticeable for SOA-iv (Figure 6), which is the dominant OA component in the southern Scandinavian region (Figure 3).

9. *Line 638-641: "Therefore, we expect that the discrepancy in this season is related to sources that are missing or underestimated in emission inventories, such as residential wood combustion in winter (Denier van der Gon et., 2015) and additional oxidation pathways" Here the important sources are clearly stated (i.e. residential wood combustion). Please add also explicitly the additional oxidation pathways that could be missing and the uncertainties in SOA yields due to wall loss in chambers.*

   Aqueous-phase and heterogeneous oxidation reactions of organics are not included in our model and can be considered as a possible cause of the OOA underestimation. This information, together with the wall losses in chambers as an additional source of uncertainty, has been added in the revised text.

10. *Line 687-689: "Nevertheless, SOA was still underpredicted during winter (NMB = -76%) indicating that other processes (e.g., seasonally dependent emissions and alternative oxidation paths) are a main cause of the inadequate performance" Also in the conclusion part, I would explicitly mention the possible underestimation of residential wood combustion emissions as a possible reason for the underprediction of SOA during winter. Please consider adding more explicitly which additional oxidation pathways could be missing and again the uncertainties in SOA yields due to wall loss in chambers.*

    We have revised the text accordingly.

References

IPCC: (Intergovernmental Panel on Climate Change): The physical science basis. Contribution of working group I to the fifth assessment report of the intergovernmental panel on climate change. T.F. Stocker, D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V.

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Jo, D. S., Park, R. J., Kim, M. J., and Spracklen, D. V.: Effects of chemical aging on global secondary organic aerosol using the volatility basis set approach, Atmos. Environ., 81, 230-244, 2013.

Lelieveld, J., Evans, J. S., Fnais, M., Giannadaki, D., and Pozzer, A.: The contribution of outdoor air pollution sources to premature mortality on a global scale, Nature, 525, 367-371, 2015.

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Zhang, Q., Jimenez, J. L., Canagaratna, M. R., Ulbrich, I. M., Ng, N. L., Worsnop, D. R., and Sun, Y. L.: Understanding atmospheric organic aerosols via factor analysis of aerosol mass spectrometry: a review, Anal. Bioanal. Chem., 401, 3045-3067, 2011.