

Replies to the Anonymous Referee 1

I am pleased to see that the manuscript has been substantially improved.

It is now of more general interest than the previous version (...). Some further comments/suggestions regarding the revised version, and the answers to my initial review are given below.

We thank the reviewer for the previous and additional comments on our manuscript. Please find below our additional responses (in black) after the referee comments (in blue). Changes in the revised manuscript are written in *italics*.

- "Additional formation of BSOA from O₃ and NO₃ is taking into account following the same approach as in Murphy and Pandis, 2009 (Menut et al., 2021; Zhang et al., 2015, 2013)."

This is still unclear to me. As far as I can see, none of the four references provide details about the SOA yields from the O₃ and NO₃-reactions of the VOCs. It is also a bit confusing to give four different references here – please stick to the one closest to your present implementation (I guess this would mean Zhang et al., 2013), in addition to the original Murphy and Pandis, 2009.

As far as I can see, the Zhang et al., 2013 study only used "low-NO_x"-yields for all SOA-formation – is this true also for the present study? Or did you use different low-NO_x and high-NO_x yields depending on the NO_x/VOC levels?

To make all these things clear, I suggest that you add a Table in the Supplement that gives the VBS-yields for the different SOA-precursors (isoprene, MT, SQT and the relevant AVOCs), and specify for which oxidants the yields are used – please also include information about low-NO_x and high-NO_x conditions in the table (if different yields are used for different conditions).

We agree with the referee, and we have now included a supplementary table to clarify and summarize all the details about the yields adopted in the VBS version of CHIMERE, i.e., Table 1 below (based on Zhang et al., 2013). Those yields applied to the whole SOA oxidants. For isoprene, only oxidation by OH are considered for BSOA formation. The differentiation of low-NO_x and high-NO_x is done as in Cholakian et al 2018 (which is based on the work of Zhang et al, 2013). Specifically, the yields of the two chemical regimes were calculated in the model with a tagged parameter (alpha) which calculates the ratio of the reaction rate of RO₂ radicals with NO (high-NO_x regime) with respect to the sum of reaction rates of the reactions with HO₂ and RO₂ (low-NO_x regime). More information is available in Cholakian et al., 2018 and Carlton et al., 2009. Additionally, throughout the whole manuscript we now refer to the study of Zhang et al 2013 as main reference for the implementation (on top of the original Murphy and Pandis, 2009).

Table 1: Mass SOA yields for each SOA precursors used in the VBS version of CHIMERE. Yields are applied to all oxidants, i.e., O₃, NO₃, and OH. For isoprene, only oxidation by OH are considered for BSOA formation. The differentiation between low-NO_x and high-NO_x yield is based on the approached proposed by (Cholakian et al., 2018).

SOA precursors	Mass yield of each bin				
	C* (µg m ³)	1	10	100	1000
ALK4	0.0	0.075	0.0	0.0	0.0
ALK5	0.0	0.300	0.0	0.0	0.0
OLE1	0.0045	0.009	0.060	0.225	0.225
OLE2	0.0225	0.435	0.129	0.375	0.375
ARO1	0.075	0.225	0.375	0.525	0.525
ARO2	0.075	0.300	0.375	0.525	0.525
TERP	0.1073	0.0918	0.3587	0.6075	0.6075
ISOP	0.009	0.03	0.015	0.000	0.000
SQT	0.0750	0.1500	0.7500	0.9000	0.9000

We added the following sentence at line 157 of page 6 of the revised manuscript as below:

The complete list of the different SOA yields used in the model are reported in Table S1.

• Q: How was the deposition of gas-phase SVOCs treated in the different model simulations? A: For this applications, SVOCs wet depositions were kept identical among all the different simulations, i.e., the model does not account for the volatility dependence of the Henry's law water solubility coefficients. We added this additional information at page 6, line 180 of the revised manuscript as below. "No volatility dependence of the Henry's law water solubility coefficients is included."

What about dry deposition? How did you treat dry deposition of gas-phase SVOCs?

The dry deposition of gases was treated with the Wesely model (Wesely, 1989). We added this information at line 182, page 6 of the revised manuscript as below:

The dry deposition of gases was treated with the Wesely scheme (Wesely, 1989)

• We additionally compared model data against organic carbon (OC) measurements as available from 15 additional EBAS sites (Table S1) and at different time resolution (from 1 day to 1 week). Since the model uses the organic aerosol (OA) mass concentration in its own calculations, we applied the OA/OC ratio as in Bergström et al., 2012.

This is unclear. Does the model track both OC and OM? If it only tracks OM – which OM/OC ratio(s) do you apply for the comparison to the OC-measurements? A single ratio for all types of OA? Or did you use different ratios for different types of SOA and POA?

We thank the reviewer for this comment. The model only tracks the OM mass. For this application we employed a single ratio for the OM/OC ratio, i.e., 1.7. We believe this is a fair approach for the comparison of model's results at rural sites during summer period. We revisited the sentence at line 316 page 11 as below:

We applied the OM/OC ratio of 1.7 as representative for biogenic secondary organic aerosol (Bergström et al., 2012).

- Extremely low OA concentrations are missed by the model (Figure 9). The latest might suggest uncertainties in the background OA fields used in the model and/or in the concentrations injected at the very boundaries of the coarser domain (i.e., long-range transport).

I think this is still confusingly formulated – do you mean that the model produces too low OA concentrations during periods with low measured OA? That is, that the model tends to underestimate OA for these periods? If so, perhaps you could write something like: The model tends to underestimate OA at Hyytiälä, especially during periods with low measured concentrations. Also, I think that the start of the second sentence “The latest” should be changed to “This”.

We agree with the reviewer, and we reformulate the sentence at line 323 page 12 of the revisited manuscript as below as suggested by the reviewer:

The model tends to underestimate OA at Hyytiälä, especially during periods with low measured concentrations (Figure 9). This might suggest uncertainties in the background OA fields used in the model and/or in the concentrations injected at the very boundaries of the coarser domain (i.e., long-range transport).

Additional technical corrections:

Line 469: CAM -> CAMS

Corrected.

Figure 10 caption; Add information that the comparison for the ebas sites is for OC: “and OC at available EBAS sites”.

Added.

Figure 12 caption: Add information that the results are for the SMEAR-II site

Added.

The Menut et al. (2021) reference should be updated to the final published version of the article (the manuscript version refers to the preprint)."

Corrected.

Reference

- Bergström, R., Denier van der Gon, H.A.C., Prévôt, A.S.H., Yttri, K.E., Simpson, D., 2012. Modelling of organic aerosols over Europe (2002–2007) using a volatility basis set (VBS) framework: application of different assumptions regarding the formation of secondary organic aerosol. *Atmos. Chem. Phys.* 12, 8499–8527. <https://doi.org/10.5194/acp-12-8499-2012>
- Carlton, A.G., Wiedinmyer, C., Kroll, J.H., 2009. A review of Secondary Organic Aerosol (SOA) formation from isoprene. *Atmos. Chem. Phys.* 9, 4987–5005. <https://doi.org/10.5194/acp-9-4987-2009>
- Cholakian, A., Beekmann, M., Colette, A., Coll, I., Siour, G., Sciare, J., Marchand, N., Couvidat, F., Pey, J., Gros, V., Sauvage, S., Michoud, V., Sellegri, K., Colomb, A., Sartelet, K., Langley DeWitt, H., Elser, M., Prévôt, A.S.H., Szidat, S., Dulac, F., 2018. Simulation of fine organic aerosols in the western Mediterranean area during the ChArMEx 2013 summer campaign. *Atmos. Chem. Phys.* 18, 7287–7312. <https://doi.org/10.5194/acp-18-7287-2018>
- Wesely, M.L., 1989. Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models. *Atmospheric Environment (1967)* 23, 1293–1304. [https://doi.org/10.1016/0004-6981\(89\)90153-4](https://doi.org/10.1016/0004-6981(89)90153-4)