

Anonymous Referee #1

The manuscript describes and evaluates a module for simulating organic aerosol, its volatility, and the oxidation state in a global model framework. The module is one of the most advanced available for global scale atmospheric models and thus the manuscript is well within the scope of Geoscientific Model Development. The manuscript is for the most part well written. However, some parts which are detailed below need clarification and correcting. I can recommend publishing the manuscript after the following issues have been considered:

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

1/ The treatment of POA is unclear to me. Do you assume some volatility distribution for it? According to Page 13 this seems to be the case as it is said that it evaporates due to dilution (Line 340). What O:C ratio or O:C ratio distribution is assumed for POA?

Similar to ORACLE v1.0, POA in ORACLE-2D is treated as semi-volatile and chemically reactive. Specific information regarding the assumed volatility distribution and O:C of freshly emitted POA for the presented model application is given in section 3.2. POA from anthropogenic fuel combustion and open biomass burning are divided based on their volatility into LVOCs (with C^* at 298 K equal to $10^{-2} \mu\text{g m}^{-3}$) SVOCs (with C^* at 298 K equal to 10^0 and $10^2 \mu\text{g m}^{-3}$) and IVOCs (with C^* at 298 K equal to 10^4 and $10^6 \mu\text{g m}^{-3}$). Their corresponding emissions are estimated using the emission factors of Tsimpidi et al. (2016). Freshly emitted POA from anthropogenic and open biomass burning sources are assigned an initial O:C of 0.1 (Figure 1a) and 0.2 (Figure 1b), respectively (Donahue et al., 2011).

2/ What is assumed for the size distribution of OA which partitions from gas to particles? Does it follow the modal approach presented in Tsimpidi et al., 2014?

Yes, the modal approach is used. The change in aerosol mass of each size mode after the phase partitioning is determined by using a weighting factor as described in Tsimpidi et al. (2014). This information has been added in section 2.2.1

3/ Page 9: All the abbreviations of organic aerosol species are not explained in this paper, i.e. aSOA-v, bSOA-v, SOA-sv, SOA-iv

Thank you for pointing this out. The same naming convention as in Tsimpidi et al.(2014) has been used. Nevertheless, for clarity, we have added an explanation of the abbreviations in the revised text.

4/ Page 10, Section 3.5: After reading this section it is still not obvious to me how gas-particle partitioning is actually calculated. Does $C_{a,i}$ in Equations (11) and (12) correspond to $SOA_{i,j}(t + \Delta t)$ in Equation (14)? If so, is it also correct that Equation (11) is first solved for one

volatility bin and then the total concentration in that bin is divided among the O:C bins using Equations (13) and (14) based on the O:C distribution on the previous time step? Do you use mole fraction from the previous time step for all the volatility bins? This procedure should be explained in more detail.

Equation 11 and 12 is a set of n nonlinear equations, where n is the number of volatility bins. Once this set of equations is solved, $C_{a,i}$ and $(C_{t,i} - C_{a,i})$ for each volatility bin i correspond to $\sum_{j=1}^m SOG_{i,j}(t + \Delta t)$ and $\sum_{j=1}^m SOA_{i,j}(t + \Delta t)$ in eqs. 13 and 14, where m is the number of O:C bins, and are used to calculate the $SOA_{i,j}(t + \Delta t)$ and $SOG_{i,j}(t + \Delta t)$ based on the O:C distribution of the previous time step. This has been made clearer in the revised manuscript.

5/ In Equation (11) it is assumed that the saturation ratio at the particle surface is dependent on the mole fraction of compounds in the organic phase. This is a common practice when solving SOA partitioning, but how good is this assumption in the atmosphere? In many cases, especially at the top of the boundary layer, humidity can be so high that the majority of moles in the aerosol is water decreasing significantly the mole fraction of individual SOA species compared to when only organic phase is taken into account. Thus SOA formation is underestimated. Can you comment on this?

We thank the reviewer for pointing this out. Indeed, water uptake by the organic-phase under high relative humidity conditions can increase the aerosol mass and decrease the mole fraction of individual SOA species which can encourage additional condensation of SOA (Jathar et al., 2016). However, this is not taken into account by ORACLE and can result in an underestimation of SOA in areas with high humidity and significant SOA concentrations. This discussion has been added to the text.

6/ The model is evaluated with observations only at the ground level. However, accounting for the volatility has implications on the vertical profiles of OA and as there are aircraft campaigns where OA has been measured with aerosol mass spectrometers, it would be interesting how well the model compares with the observations in the vertical.

This is a very interesting suggestion. We believe that such a comparison can provide significant insights on the vertical profile of OA and its properties (e.g., hygroscopicity) due to the chemical aging and changes in its volatility and oxidation state. Therefore, we believe that such an analysis will be more suitable in a separate manuscript since the main scope of the current work is to present the new submodel, its capabilities and its technical implementation.

7/ Can you give some estimate on the increase in the computation time when moving from the 1-D ORACLE to 2-D ORACLE?

Similar to ORACLE, ORACLE 2-D has a flexible interface in which the user can choose the resolution (number of bins used in each dimension) of the 2-D VBS space through a namelist file depending on the desired application and scientific goals. The chemical resolution employed here includes 164 organic aerosol surrogate compounds, compared to 34 OA compounds in the original ORACLE paper (Tsimpidi et al., 2014), which result in a 16% increase of the EMAC computational burden. This is discussed in section 3.1

Technical comments and typos:

1/ Page 2, Line 36: space missing before the word “Organic”

Corrected.

2/ Page 3, Line 44: pas-aerosol → gas-aerosol

Corrected.

3/ Page 10, Line 270: equation → equations

Corrected.

4/ Figure 1a: Compustion → Combustion

Corrected.

5/ Figure 8 caption: To be consistent with the rest of the manuscript, please change SV-OOA → SVOOA and LV-OOA → LVOOA

Done.

References

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- Jathar, S. H., Mahmud, A., Barsanti, K. C., Asher, W. E., Pankow, J. F., and Kleeman, M. J.: Water uptake by organic aerosol and its influence on gas/particle partitioning of secondary organic aerosol in the United States, *Atmospheric Environment*, 129, 142-154, 10.1016/j.atmosenv.2016.01.001, 2016.
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