Authors' response to comments made by anonymous reviewer #1:

Summary

This work documents a newly-developed model (ORACLE-lite) for organic aerosol with simplified representation of organic aerosol volatility and aging chemistry and embedded as a module in the global model ECHAM/MESSy, and further demonstrates the model's ability to improve computational efficiency (in comparison to a previous version of the ORACLE model) and to capture observed OA/POA/SOA concentrations across the globe. Several model deficiencies demanding further strengthening are identified (e.g., aqueous SOA formation). Overall, this works represents a concrete step of advancement of the ECHAM/MESSy-ORACLE modeling system, which has continuously progressed over the past decade. There is no major issue from this reviewer and publication is recommended if the following comments can be addressed.

We thank the reviewer for the positive assessment of our work and for recognizing ORACLE-lite as a concrete advancement of the ORACLE family modeling framework within the EMAC system. Below is a point-by-point response (in black) to the reviewer's comments and suggestions (in blue).

Comments

1. *In Figure 1, change "Sort Term" to "Short Term"*

Thank you for noting this typo. "Sort Term" has been corrected to "Short Term" in Figure 1.

2. *In Figure 2, the meaning of SOA-sv/SOA-iv/SOA-v are not immediately clear; would be helpful to add a legend.*

Thank you for this suggestion. We have revised Figure 2 to explicitly define SOA-sv, SOA-iv, and SOA-v as secondary organic aerosol formed from SVOC, IVOC, and VOC oxidation, respectively.

3. *In Line 190-191, the authors used the term "emission factor", which is confusing, because what the authors really mean is the emission ratio of I/S/LVOCs to the traditional emission factor of POA. Please revise the text here.*

We agree that the term "emission factor" was misleading because the values represent the ratios of LVOC, SVOC, and IVOC emissions relative to traditional POA emissions. We have therefore replaced "emission factor" with "emission ratio" and revised the corresponding discussion to clarify how these ratios are applied, including the treatment of additional IVOC emissions from anthropogenic combustion sources.

4. *In Line 235, it is stated that "only homogeneous gas-phase aging is included". Since the ECHEM-MESSy-ORACLE system is intended for long-term global simulations, wouldn't heterogeneous oxidation have a substantial impact on this spatial and temporal scale (Hodzic*

et al., 2016)? If the authors agree, please acknowledge this as an issue for future improvement in the Conclusions Section.

We thank the reviewer for raising this important point. We agree that heterogeneous oxidation represents an additional OA aging pathway that is not currently included in ORACLE-lite and that it may influence OA evolution over sufficiently long atmospheric timescales (Hodzic et al., 2016) (Hodzic et al., 2016). At the same time, its relative importance depends on OA volatility and gas–particle partitioning. For semivolatile organic compounds, gas-phase oxidation is substantially faster than heterogeneous oxidation and is therefore expected to dominate their oxidative aging (Donahue et al., 2013). Heterogeneous oxidation may become relatively more important for LVOCs that reside predominantly in the particle phase, although heterogeneous processing generally occurs over longer timescales of several days (Kroll et al., 2015). In addition, the phase state of OA can introduce kinetic limitations to condensed-phase processing, particularly in the middle and upper troposphere where SOA is predicted to be predominantly glassy (Shiraiwa et al., 2017). We therefore consider the present gas-phase aging treatment an appropriate simplification for capturing the large-scale OA evolution targeted by ORACLE-lite, while recognizing that heterogeneous oxidation represents an additional process that could be considered in future model improvements. We have added this point to the Conclusions.

5. *In Figure 4a, there seems to be some hotspots of OA in Canada. Are there any justifications for these hotspots? There seems to be no discussion of it in the current text.*

The POA hotspots over Canada are associated with biomass-burning emissions from boreal forest fires. The high primary organic emissions in these regions, combined with the enhanced partitioning of emitted semivolatile material into the particle phase in ORACLE-lite, result in elevated POA concentrations. This is also reflected in the positive differences relative to ORACLE-base shown in Figure 4b. We have added a brief explanation of these hotspots to the discussion of Figure 4.

6. *In Figure 4b, the OA prediction from ORACLE-lite differs significantly from the prediction from ORACLE-base, mainly due to a change of volatility bins used to represent POA. Since it is not definite which representation (ORACLE-lite or ORACLE-base) is better, which results should be trusted more? Please elaborate and add to the text.*

We thank the reviewer for this important comment. ORACLE-base employs a larger number of surrogate species and a higher volatility resolution and therefore provides, in principle, a more detailed representation of gas–particle partitioning. The differences shown in Figure 4b are a direct consequence of the reduced volatility resolution in ORACLE-lite, which enhances the partitioning of freshly emitted semivolatile material into the particle phase and consequently leads to higher POA concentrations in some regions. This behavior represents a necessary compromise associated with the reduced-complexity design of ORACLE-lite, which substantially decreases the computational cost and enables long-term global chemistry–climate simulations. However, the suitability of this simplification is independently assessed through the extensive comparison with AMS and ACSM observations presented in Section 3.2. Despite its reduced volatility resolution, ORACLE-lite reproduces the observed magnitude, spatial variability, and major compositional

features of OA reasonably well across diverse regions and seasons. Thus, while ORACLE-base retains a more detailed representation of the OA volatility distribution, the observational evaluation demonstrates that ORACLE-lite maintains sufficient accuracy for the long-term global applications targeted in this study. We have clarified this in the revised manuscript.

7. *In Section 3.2.1, there lacks a description of the modeling configuration; that is, what emission inventories and meteorological fields (if any) are used. Are the emission inventories up-to-date?*

We thank the reviewer for pointing out that the description of the simulation setup was incomplete. We now specify in Section 2.1 the emission inventories used for the different source categories and the meteorological fields used to constrain the simulation.

8. *In Section 3.2.1, it is stated that "Overall, the model reproduces the large-scale spatial variability of OA across regions, with most locations showing agreement within a factor of two", which is not substantiated by the data shown in Figure 6 alone. Therefore, the reviewer suggests that Figure 6 and Figure 7 be combined to form a single Figure, so that the scatter plots can be visible alongside the relevant text.*

We thank the reviewer for this helpful suggestion. We agree that the statement regarding agreement within a factor of two is more directly supported by the scatter plots than by the spatial deviations shown in Figure 6 alone. We have therefore combined the former Figures 6 and 7 into a single figure, allowing the spatial distribution of model deviations and the corresponding model–observation comparison to be viewed together.

9. *In Section 3.2.1, it is not clear whether the model-measurement comparison is on basis of annual average or 20-year average. Please clarify early in this section.*

We thank the reviewer for pointing out that this was not sufficiently clear. For each AMS/ACSM dataset, the simulated concentrations are sampled at the corresponding measurement location and averaged over the respective measurement period. We have clarified this in the new Section 2.4.

10. *In Line 523, "key missing SOA formation pathways" is mentioned; thus, a comprehensive discussion of the missing pathways should include auto-oxidation of larger molecules (e.g., monoterpenes) to rapidly form extremely low volatility organic compounds (Bianchi et al., 2019).*

We agree that autoxidation represents an additional SOA formation pathway that is not explicitly represented in ORACLE-lite. In particular, autoxidation of larger VOCs, such as monoterpenes, can rapidly produce highly oxygenated organic molecules with very low or extremely low volatility, thereby contributing to SOA formation (Bianchi et al., 2019). We have added this process to the discussion of missing SOA formation pathways.

11. In Line 99, the ORACLE-lite model is described as "standalone". If so, would it be valuable for the OA model to be configured as a 0-dimension box model to simulate laboratory experiments in the literature to further validate model predictions in an isolated setting? Please consider this possibility and discuss it in the Conclusions Section.

We thank the reviewer for this valuable suggestion. We agree that ORACLE-lite could also be applied in a 0-D box-model framework for process-level evaluation against laboratory chamber experiments under controlled conditions. The formulation of ORACLE-lite allows the OA chemistry and gas–particle partitioning calculations to be applied independently of the full global model, provided that the relevant experimental conditions and chamber-specific processes are prescribed. Such an application could provide a complementary approach for evaluating individual model processes and parameterizations. We have added a brief discussion of this potential application in the Conclusions.

References

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