We thank the reviewer for his/her comments. Here they are reported (in bold) with our replies.

1. line 16: use "secondary organic aerosols" instead of just "aerosols", because it is already said that the aerosols discussed here are from VOCs.
   We have applied the change.

2. line 37: Can the authors elaborate in the manuscript why explicit chemical identities would be useful in atmospheric simulations here?
   Explicit chemistry schemes allow a representation of Secondary Organic Aerosols formation from VOCs without the use of lumped species and experimentally derived parameters (e.g., reaction rates, aerosol yields). These tuning parameters can increase model uncertainties and result in large differences between Atmospheric Chemistry models. In addition, empirical chemical schemes are not mass conserving (e.g., for carbon) and the higher-generation reaction products are lumped or ignored, even if, for instance, they play a pivotal role for OH recycling and ozone chemistry (Taraborrelli et al., 2012), or are a major component of atmospheric brown carbon (Laskin et al., 2015). In general, explicit identities of model species are essential for making further progress in quantifying the atmospheric budget of Secondary Organic Aerosols. By relying on high-quality (experimental and theoretical) data of the physico-chemical properties of precursors and intermediates, an ever more realistic modelling of removal, ageing and formation pathways would be possible.

3. For the OA treatment (starting from line 103), has the MOM + ORACLE approach been tested against chamber SOA experiments of, for example, alpha-pinene, to see if the model correctly captures SOA mass production in the experiments? This reviewer trusts that the two submodels have been well tested in previous studies but just wonders if this can be done for closure.
   This is a very important aspect to look at in order to ensure an ever increasing realism of our model as experimental capabilities and knowledge progress. Beside the global model, MOM is normally run in the photochemical box model CAABA/MECCA (Sander et al., 2019), which allows for multi-phase kinetics simulations with deliquescent aerosols and cloud droplets, and it has been evaluated with chamber studies (e.g., Novelli et al., 2020). Nevertheless, the MOM + ORACLE approach has so far not been evaluated against chamber experiments, but this is planned for modelling the experiments routinely done at the SAPHIR and SAPHIR* chambers (https://www.fz-juelich.de/iek/iek-8/EN/Expertise/Infrastructure/Infrastructure_node.html). However, the MOM + ORACLE approach has been evaluated against observation with the mixed layer model by Janssen et al. (2017).

4. For section 4.2.2, the authors do not seem to offer a potential explanation for the underestimation. Can this be added to the text?
   It is indeed difficult to assess a potential explanation for the strong and homogeneous underestimation. Due to their lower solubility, these tracers are only removed by reactions with OH, NO3, and O3, with the first being at least two orders of magnitude faster than the other. There are therefore strong indications that the reaction with OH of these tracers are too fast. Analogously to Pozzer et al. (2006), we again point to a revision of such reaction rates. Furthermore, beside possibly too fast decomposition of these tracers, a substantial lack of emissions could be present, even from natural sources as shown by Li et al. (2021).

5. In the outlook section, the authors discuss potential improvements to the model.
Since SOA lifecycle in the atmosphere can also be affected by aerosol phase state, heterogeneous reaction with oxidant, etc., which are still highly uncertain, could the authors include these processes in the discussion as well, and talk about how they can be captured in the MOM+ORACLE framework?

The referee pointed out correctly some further issues that could be implemented in the future. Some of them have already been studied or implemented in the EMAC model, although not explicitly described or used here.

Initial work on the state of the secondary organic aerosols with the ORACLE model has already been performed by Shiraiwa et al. (2017), although more work is needed to estimate its impact on the global secondary organic aerosol budget. The MOM+ORACLE framework calculates the phase partitioning of organic compounds by assuming a bulk equilibrium. However, the phase state of the organic aerosol can affect the mixing time of the condensed organic compounds within the aerosol. In general, equilibrium partitioning to the particle phase is a reasonable assumption if the aerosol is liquid, however, if the phase state is solid, non-equilibrium partitioning should be considered. The MOM+ORACLE framework does not consider any kinetic limitations in the bulk. Shiraiwa et al. (2017) suggests that kinetic limitations in the bulk may not significantly affect SOA partitioning in the boundary layer, justifying the use of equilibrium partitioning in this part of the atmosphere.

Finally, MOM comprises an explicit gas-phase VOC oxidation scheme but does not include any heterogeneous reactions. However, the MOM+ORACLE framework can be used as a basis to link ORACLE with an aqueous phase mechanism which is planned for a future study.

References


