Responses to the Comments of the Reviewers

Reviewer 1

General comments

(1) The manuscript "Implementation of the ORACLE (v1.0) organic aerosol composition and evolution module into the EC-Earth3-AerChem model" by Kakavas et al. describes briefly the implementation ORACLE (v1.0) organic aerosol model in TM5-MP atmospheric chemistry model. It is analyzed and evaluated using it as a standalone model driven with reanalysis data as well as coupled to EC-Earth3-AerChem Earth system model. The manuscript fits in the scope of the journal addressing the need for a computationally efficient and scientifically accurate description for organic aerosol. It presents a novel implementation of ORACLE-lite module into TM5-MP which shows improvements in simulated OA concentrations in both configurations it is evaluated. Methods are scientifically valid, although the manuscript relies on its description a lot on previous papers published on ORACLE-lite. However, for example Pandis et al. (1993) paper is behind a pay wall. The results mainly support the interpretations and conclusions. The description could be more detailed so that the reader shouldn't have to go through so much of previous publications on ORACLE. The paper is well structured, the language is fluent and precise.

We appreciate the positive assessment of our work by the reviewer. We have tried to address all comments of the reviewer and to improve the paper accordingly. We have followed the suggestion of the reviewer and added a more detailed description of the ORACLE module in the corresponding section of the manuscript. Our responses (in black) and the corresponding changes in the manuscript follow each comment of the reviewer (in blue).

Specific comments

(2) Line 88: "ORACLE reduces the computational cost by utilizing a small number of surrogate OA species by employing a novel lumping method.". When compared to what?

We have rewritten the sentence to clarify the basis of the comparison.

(3) Line 116: There is a description paper by van Noije et al., (2023) on this EC-Earth3-Aerchem which might be a more appropriate reference for this model. In addition, Döscher citation is for year 2021, but the reference is 2022.

We have followed the reviewer's suggestion by adding the reference to van Noije et al. (2021) and correcting the Döscher citation year to 2022.

(4) Lines 180-186: "Note however, that the SOA formation from biogenic VOC emissions (isoprene and monoterpenes) is already represented in the models, as described by Bergman et al. (2022), while SOA formation from anthropogenic VOC emissions is neglected. As a result, the number of surrogate species used to represent OA and its volatility in ORACLE-lite was reduced from 18 to 9. An overview of the characteristics of the lite configuration of the ORACLE module used in this study is shown in Table 1." This is unclear what is meant by this. Is SOA formation from biogenic VOC emissions treated separately using the approach by Bergman et al. (2022)?

This is correct. SOA formation from biogenic VOC emissions is treated separately in the model using the approach described by Bergman et al. (2022). To clarify this point and avoid confusion, we have added a brief discussion in this section of the revised manuscript.

(5) It is also unclear how OA is partitioned between different modes. Pandis et al., (1993) is referred to, but I don't have the access to the paper.
We have added a brief discussion in this section of the revised manuscript explaining how OA is partitioned among the different modes.

(6) Are SOA species affecting aerosol radiative properties? If so, what assumptions have been used for optical properties?
The formed SOA does interact with radiation. We have added a brief discussion of the assumed optical properties in the revised manuscript to clarify this aspect.

(7) Do SOA species affect cloud droplet activation in EC-Earth?
Yes, they do. SOA contributes to the organic aerosol mass within the modal aerosol scheme (M7), so it affects aerosol growth and particle properties. As cloud droplet activation depends on aerosol size, number, and hygroscopicity, SOA indirectly influences cloud droplet activation in the model through changes in both the aerosol size distribution and composition. We have added a brief discussion in the revised manuscript.

(8) Line 231: "data were available from only 3 stations for the simulated period." Could you have used data for another year or ran a year that would have more station data?
Following the reviewer's suggestion, we have extended the evaluation of the results of the online simulation of EC-Earth3-AerChem for 2010, for which more observational stations are available within the EMEP framework.

(9) Page 9: Emissions between TM5-MP and EC-Earth look remarkably similar. Is there a good reason to show both of them? It would also be good to use some other colormap and scale, and put them in the same figure to see if there are any differences. Jet colormap is also not recommended in modern data visualization. The same comment applies to Figure 3.
TM5-MP and EC-Earth simulations use the same emissions. The TM5-MP emissions shown in Fig. 1 correspond to the year 2005, whereas the EC-Earth3-AerChem emissions shown in Fig. 2 represent the annual mean over 2000–2010. Following the reviewer's suggestion, we have combined Figs. 1 and 2 into a single figure and replaced the current colormap with a more appropriate one.

(10) Lines 456-458: "The incorporation of ORACLE-lite significantly improved the representation of OA formation and atmospheric behavior both in the standalone TM5-MP and the EC-Earth." This is an inaccurate sentence since the default TM5-MP does not simulate OA formation, rather they emit OA as primary particles. In addition, overall significance of improvement depends on which evaluation metrics we are looking at in Table 3. Bias shows clear decrease, but different error metrics not necessarily.

We have followed the suggestion of the reviewer and changed this sentence to "The incorporation of ORACLE-lite reduced the bias of the OA predictions both in the offline and online simulations of EC-Earth3-AerChem.".

(11) Lines 477-479: "The seasonal and spatial variability of SOA was also better captured, with higher concentrations predicted in regions with intense biomass burning and anthropogenic activity, such as India, China, and sub-Saharan Africa." How do you determine better seasonal and spatial variability? To me, these are not evident from the manuscript.
We have removed this sentence from the revised manuscript.

Reviewer 2

General comments

(1) The current work provides a description of the implementation of the ORACLE-lite secondary organic aerosol (SOA) formation mechanism in the TM5-MP and EC-Earth modeling systems. Accurate modeling of SOA concentrations is important for both air quality and climate purposes, even though SOA modeling is often associated with considerable uncertainty and computational cost. The implementation and evaluation described in the current work serves as a useful source of documentation, both for the currently described models and for other models that may wish to implement the ORACLE-lite module in the future. The comparison against surface observations, while limited to North America and Western Europe, is an important element of this work. I can recommend this article for publication after the below comments have been considered.

We appreciate the positive assessment of our work by the reviewer. We have tried to address all comments and to improve the paper accordingly. Our responses (in black) and the corresponding changes in the manuscript follow each comment of the reviewer (in blue).

(2) The authors note that only 3 stations from the EBAS database are available for the year 2005. However, following the link provided to the EBAS database finds that PM2.5 OC measurements are available from 4 stations (one in Spain, Italy, Germany, and Norway). The Italian, German, and Norwegian sites all have nearly continuous measurements over the course of the year. The Spanish site only has two daily mean measurements per month. Given that it is typical to calculate monthly means only when around 60-70% data availability is reached, I think the measurements from the Spanish site should be excluded from the analysis. At least so long as the analysis focuses on monthly mean values. The Italian site (Ispra) is clearly influenced by anthropogenic wood burning emissions, reaching a monthly mean OC concentration of 22 $\mu g/m^3$ in January (or 40 $\mu g/m^3$ OM using the employed OA/OC ratio of 1.8). This value would skew the average monthly mean across all sites (whether the Spanish site is excluded or not) beyond the average of 2.2 $\mu g/m^3$ currently shown for January in Figure 8. It therefore seems like the Italian site was excluded from the analysis. However, since this is a rural site (as are the German and Norwegian sites), and since the high wintertime concentrations are likely to have large contributions from the anthropogenic biomass burning emissions (and the resulting oxygenated OA formation) that is central to the current work, it is not clear to me why this station should be excluded. The availability of OC measurements in Europe increases greatly in later years. For example, for 2010 there are 8 stations with good data availability, whereas for 2015 there are 20. I think the simulations should at least be compared to EBAS/EMEP observations for the year 2010 (implying that a new simulation with TM5-MP would have to be performed). Ideally, however, more recent years for both the TM5-MP and EC-Earth simulations would have to be considered, preferably for 2015 or later. If the authors decide to continue using the TM5-MP simulation for 2005, I think the stations in Germany (Melpitz), Italy (Ispra), and Norway (Birkenes) should be evaluated in detail on an individual basis rather than as the average across all three sites.

This is a valid point. The OC measurements at the Ispra (Italy) station are systematically high (reaching up to 22 $\mu g\ m^{-3}$ in winter), which strongly influences the multi-site monthly mean shown in Fig. 8 due to the limited number of available stations for 2005. For this reason, and to avoid the average being dominated by a single site, Ispra was excluded from the statistical analysis. This is now clarified in the revised manuscript. Also, we have followed the suggestion of the reviewer and added the evaluation of EC-Earth3-AerChem for the year 2010 in the revised manuscript, when more observational stations are available. Since the differences in predicted OA concentrations between TM5-MP and EC-Earth are relatively small, we consider that performing an additional TM5-MP simulation for 2010 is not necessary for the purposes of this comparison. A brief discussion has been added in the corresponding section of the manuscript.

Specific comments

(3) It would be helpful if the ORACLE-lite description would be expanded to make the article more self-contained, even though more detailed descriptions can be found in the cited works. For example, it would be helpful if the K_OH rates that modify the volatility of the emitted organic compounds are included in Table 1.
We have followed the suggestion of the reviewer and expanded the description of ORACLE-lite to make the manuscript more self-contained and added the corresponding reaction rate constants of SVOCs and IVOCs with OH to the revised manuscript.

(4) Could the motivation behind neglecting SOA formation from anthropogenic VOC emissions be expanded? These are said to contribute only 15% to total global average surface OA concentrations, but this seems like a considerable amount (similar also to the calculated annual SOA-sv mass, roughly based on Table S1). The measurements are also underestimated by 8-13% with the current ORACLE-lite setup, suggesting that the additional SOA from VOCs would improve/reduce the model bias. The current runtime increase of 8% seems modest. How much slower would the model become with the addition of SOA from anthropogenic VOCs?
This is a point that deserves additional discussion. Although the additional SOA formed from anthropogenic VOCs could improve the model bias, ORACLE-lite was originally developed for use with the SAPRC family of gas-phase mechanisms. In contrast, TM5-MP and EC-Earth3-AerChem employ a modified CB05 chemical mechanism, which uses a different lumping structure for anthropogenic VOCs than that assumed in ORACLE. As a result, the direct inclusion of anthropogenic SOA formation within the current ORACLE-lite framework is complex and requires additional development. This will be the topic of future work. This point is now explained in the revised manuscript.

(5) When the IMPROVE network is introduced, it would be helpful if it is mentioned that the main purpose of the network is to measure aerosols in remote areas of the United States, with the measurements therefore being representative of rural conditions.
We have followed the reviewer's suggestion and clarified in the revised manuscript that the IMPROVE network measures aerosols in remote areas of the United States and as a result is representative of rural conditions.

(6) The authors mention that measurements of OC are converted to OA using an OA/OC ratio of 1.8, while the traditional model setup is to assume that OA from all emission sources (treated as POA) has a ratio of 1.6. For reference, it would be helpful to include a description of the OA/OC ratios calculated/assumed using ORACLE-lite, and the resulting calculated ratios of the total OA.

In the present application, SVOCs and IVOCs undergo up to two generations of oxidation, with a 22.5% mass increase in each generation. Assuming an initial OM/OC ratio of 1.2 in ORACLE-lite, this leads to a final OM/OC ratio of up to 1.8, which is within the observed range for oxygenated organic aerosol (OM/OC: 1.8–2.4; Aiken et al., 2008). This is now explained in the revised manuscript.

(7) In Fig. S13, could the y-axis be changed to show hPa rather than model level? Currently it is difficult to make out the vertical scales.

We have followed the reviewer's suggestion and changed the y-axis in Fig. S13 to show pressure (hPa) instead of model levels in the revised manuscript.

(8) ECMWF is defined on both lines 120 and 131.

The duplicate definition of ECMWF in line 131 has been removed in the revised manuscript.

(9) SOA-sv is said to correspond to a C* value of 10^{-2} μg/m^3 on line 332. However, Table 1 shows a value of 10^1 ug/m3.

This is a misunderstanding. The value in Table 1 refers to the initial representative volatility bin of the emitted species, not to the volatility after aging. We have clarified this point in Table 1 of the revised manuscript to avoid confusion.

References

Aiken, A. C., DeCarlo, P. F., Kroll, J. H., Worsnop, D. R., Huffman, J. A., Docherty, K. S., Ulbrich, I. M., Mohr, C., Kimmel, J. R., Sueper, D., Sun, Y., Zhang, Q., Trimborn, A., Northway, M., Ziemann, P. J., Canagaratna, M. R., Onasch, T. B., Alfarra, M. R., Prevot, A. S. H., Dommen, J., Duplissy, J., Metzger, A., Baltensperger, U., and Jimenez, J. L.: O/C and OM/OC ratios of primary, secondary, and ambient organic aerosols with high-resolution time-of-flight aerosol mass spectrometry, Environ. Sci. Technol., 42, 4478–4485, doi: 10.1021/es703009q, 2008.

Bergman, T., Makkonen, R., Schrödner, R., Swietlicki, E., Phillips, V. T. J., Le Sager, P., and van Noije, T.: Description and evaluation of a secondary organic aerosol and new particle formation scheme within TM5-MP v1.2, Geosci. Model Dev., 15, 683–713, doi: 10.5194/gmd-15-683-2022, 2022.

van Noije, T., Bergman, T., Le Sager, P., O'Donnell, D., Makkonen, R., Gonçalves-Ageitos, M., Döscher, R., Fladrich, U., von Hardenberg, J., Keskinen, J.-P., Korhonen, H., Laakso, A., Myriokefalitakis, S., Ollinaho, P., Pérez García-Pando, C., Reerink, T., Schrödner, R., Wyser, K., and Yang, S.: EC-Earth3-AerChem: a global climate model with interactive aerosols and atmospheric chemistry participating in CMIP6 , Geosci. Model Dev., 14, 5637–5668, doi: 10.5194/gmd-14-5637-2021, 2021.