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Interactive comment on "ORACLE: a module for the description of ORganic Aerosol Composition and Evolution in the atmosphere" *by* A. P. Tsimpidi et al.

Anonymous Referee #2

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The paper by Tsimpidi et al. describes the organic aerosol module ORACLE, coupled with the global model EMAC, which takes into account the semi-volatility of all organic aerosols, both primary and secondary, from anthropogenic and biogenic sources, including the IVOCs. The module is flexible in design, both with regard to the amount of volatility bins used, and the processes included, like aging. The paper is very clearly written and the module is thoroughly documented. I recommend publication after addressing the following minor points.

Minor comments:

Although the model includes aerosol microphysics that take into account the aerosol

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hygroscopicity, no discussion is made on size-resolved and hygroscopicity results. I understand that this might be outside the scope of the paper, especially if evaluation has to come into play, but some basic discussion is missing. This includes questions like: which mode has most of the aerosol mass? Which species condenses the most in each mode? How do organic aerosols affect the hygroscopicity of the modes? What are the assumptions made for the hygroscopicity of each volatility bin? Does hygroscopicity play a role on the size-dependence partitioning described in section 3.6? Is there any link (in the model) between hygroscopicity and volatility?

Abstract, line 18: "domain-average" refers to which domain?

p. 5470, l. 20-21: One year of spinup is probably not enough for semi-volatile organics that tend to accumulate in the upper layers of the troposphere where temperatures are very low, and their lifetime is higher since they are above clouds. This can be tested by looking at the organics optical depth, or upper tropospheric burden, as a function of time.

p. 5471: please add a sentence/reference or two regarding the aqueous formation of sulfate, and, if any, organics.

p. 5473, l. 15-16: Isn't 1e-1 too volatile for ELVOCs?

Section 3.4: are there primary marine organic emissions in the model? Do you have a reference for the ONLEM submodel? What was the impact of the inclusion of aVOCs to the model's gas-phase chemistry? Why not use RCP emissions for combustion, for consistency, and use the AeroCom ones instead? Also, technically speaking, the RCP emissions are not IPCC, but CMIP5.

p. 5476, l. 10-15: This approach has limitations, since e.g. aVOCs are also emitted by biomass burning.

p. 5476, l. 20: where does the 7.5% mass increase is based?

p. 5477, l. 1: why this is not the case for all other OA?

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The aging parameterization appears inconsistent to me. bSOA do not age, aSOA age by steps of a factor of 10, and all others age by steps of a factor of 100. Are there chemical (or other) evidence for that? The enthalpy of vaporization has a similar inconsistency, why not use an enthalpy of vaporization as a function of the volatility for all species?

What molecular weights have you used for each volatility bin?

Results: Frequently Congo is mentioned, when the maximum is much wider than the country of Congo. In addition there is a strong seasonal cycle, from Sahel to southern Africa, which is not clearly visible in the annual mean, but needs to be mentioned.

p. 5481, l. 21-22: In my opinion, missing processes add primarily to the model bias, not the model uncertainty, since there is a missing source or sink. The uncertainty of the missing parameterization is second order.

Why not exclude Ispra completely from the analysis, since the model is not able to capture the unique characteristics of the station? In any case, even if Ispra stays in the analysis, how do the statistics change in case Ispra is dropped?

Sections 4.3-4.7: when mentioning global average surface concentrations, it would be useful to also mention the mean over land only, since most of the aerosols are there. You can also add these numbers in table 7.

Section 4.3, tPOA: The discussion for cities is laid in a way that might give the wrong impression that you can actually resolve them. Use something like “the greater Beijing area” or “the gridbox that includes Beijing” or similar when it comes to large urban centers.

p. 5486, l. 1-2: This is very interesting, are there any measurements that support it?

p. 5489, l. 25-27: There are studies that have challenged this statement in the past, e.g. the work of Spracklen et al.

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Figure 2: The aging arrows for SOA-v only apply to aVOCs, not bVOCs, it should be clearly mentioned. The legend should say “Gas/Particle Partitioning” not “Partition”. “Rhombi”, although literally correct, is better to be “diamonds”, since this is the symbol we are interested in the figure, not the geometrical shape. You should explain what the circles are, and probably add arrows showing which species can be emitted. Lastly, the footnote with the star should also be added at the bottom of p. 5473.

Technical corrections:

p. 5467, l. 12: “aerosol related” needs a hyphen.

p. 5468, l. 11: “logarithmically spaced” needs a hyphen.

p. 5469, l. 8: “two product” needs a hyphen.

p. 5469, l. 12: “most cases” should be “some cases”. In the next line, “etc.” is not needed: Pye lacks aging, Farina lacks semivolatiles; is there anything else included in “etc.”?

p. 5470, l. 24: NO2 is not an oxidant, maybe you mean NO3 radical?

p. 5471, l. 9: It appears there is something missing here: “same size range” with their hydrophilic counterparts?

p. 5472, l. 4: Please add “as described in section 3.6” (or something like that) after “size modes”.

p. 5472, l. 8: “high number”: please add the exact number under the present configuration.

p. 5473, l. 13: Please change “groups with” with “groups, each with”.

p. 5473, l. 19: “exist exclusively” should be “exist almost exclusively”.

p. 5474, l. 27: The range from 0.01 to 100 is elsewhere mentioned as 0.1-10, including Fig. 1. Same for the 1e3-1e6 range 2 lines later, which is mentioned 1e3-1e5

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elsewhere (including Fig. 1).

p. 5475, l. 25: “monoterpene” should be “monoterpenes”.

Eq. R8/10/12/14: The indices should be i-1, or the indices in R6 should be i. However, none of these reactions are needed here, since they show the partitioning, which is described in the following section. In any case, they are rather trivial reactions which are nicely described in the text, so they can be omitted. Line 3 from the following page also has a similar issue, it should be i-1 produces i-2, unless you change reaction R6. Lastly, this sentence (in line 3, p. 5478) should end by “until they reach the lowest volatility bin”.

The first sentence of section 4.3 is repetitive and is not needed.

p. 5485, l. 17: “more chemically processed” compared to which?

p. 5485, l. 19-20: “4 times higher than of SVOC (Table 4)”: I don’t see that in the table, but it would be good to have it there, please add it.

p. 5485, l. 26: “if” should be “though”.

p. 5488, l. 27: Where exactly is the “higher up” that has 92% of SOA?

Table 1: POG comes from direct emissions, or only from evaporation of emitted POA?

Table 4: Is the factor 2.5 already applied in the numbers in the last two columns, or it is applied on these numbers? Also, can you split the last column into the individual contributions of the S/IVOC?

Table 6: The RMSE is not discussed at all in the manuscript. Either mention it, or drop it, don’t simply include the numbers in the table.

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