Geoscientific
Model Development
Discussions

Interactive comment on "ORACLE 2-D (v2.0): An efficient module to compute the volatility and oxygen content of organic aerosol with a global chemistry – climate model" *by* Alexandra P. Tsimpidi et al.

Anonymous Referee #2

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The manuscript by Tsimpidi et al. presents the extension of the ORACLE module in the EMAC global model to include the 2-D VBS for the calculation of organic aerosol evolution in the atmosphere. It is an improvement over the previous ORACLE module, developed by the same group, which included the 1-D VBS. The paper is very well written and contains a lot of information that are useful both to model developers that want to reproduce the results but also to the audience interested in the science output of the new model. I especially want to applaud the authors for providing tables with all model results, instead of just scatterplots, which makes the comparison of their work

against future studies trivial. I also liked Figure 7 a lot; I found it very informative and interesting. I am listing a number of major and minor points below, but I do expect that after sufficient revision the manuscript will be eventually published in GMD.

Major points

Lines 79-80: While it is great that this capability exists, it has not been detailed in the manuscript, and it is expected for a model development paper. How easy is it to change the mechanism? The technical details discussed later (e.g. mass yield distribution of emissions in C* and O:C bins, photochemical aging rate and products) will be adjusted automatically using some kind of interpolation or other internal model assumption, or the user needs to start from the beginning and introduce new assumptions for any change in the complexity? Has any other complexity been tested in the model, and if yes, how does it perform against the one presented? Is the one presented the optimal one after testing several configurations, or it is the first guess?

The fragmentation discussion has a number of issues, which, in my opinion, will eventually lead to overestimation of OA in the model. First of all, the manuscript (line 233-234) states that volatility increases with fragmentation, which is not shown in figure 1. The net average decrease of volatility does not justify the elimination of the higher volatility product by simply adjusting the volatility of the low volatility product to higher values. This might work on field campaigns where over the measuring site the system remains open in terms of mass balance, but in a global model where mass conservation is paramount, a fraction of OA must leave the VBS framework as volatile compounds (e.g. CO, CO2, HCHO, acetone). If not, the carbon that is not lost, will eventually oxidize and form OA later, contributing to the total OA mass and leading to OA overestimation. This carbon that was not lost will also affect the O:C ratio of the resulted OA, by probably resulting in a net (small) decrease of O:C at areas where fragmentation is important. Overestimating OA because of the additional fragmented carbon taken into account is also important for the partitioning of the semi-volatile OA: when more OA is present in the aerosol phase, partitioning of semi-volatile compounds will move

towards the particulate phase, further adding to the OA overestimation.

The addition of oxygen to the molecules is a statistical approach that lacks chemical intuition. Every OH oxidation step, especially at the first oxidation stages, should add one O atom. If two or three are to be added, this is most likely because of isomerization reactions, which requires a long-chain VOC to be the compound being oxidized, and there is no reason for the reaction rate to be faster (as described in section 3.4). Alternatively, more than one O atoms can be added if the reactant is an unsaturated VOC, but then the reaction rate can be an order of magnitude faster, at least. I understand that for simplicity certain choices have been made, but since the model is aware of the number of carbon atoms, I believe that this information can be used in that respect.

The temperature dependence on partitioning is completely absent in the manuscipt. Its treatment though can have a huge impact on results, and should not only be presented, but also discussed in detail. Assuming that an enthalpy of vaporization is used in the model, is that constant across the two dimensions of the VBS framework, or it is changing (and how and why)? A high enthalpy of vaporization will make the volatility bins with low C* very non-volatile when temperature drops even slightly, completely shutting down gas-phase oxidation, due to the absence of gas-phase material, affecting all results presented in the manuscript.

Minor points

Bottom of page 3: It would be useful to put ORACLE v1.0 in perspective there as well.

Lines 140-143: Although already published, since it is highly relevant here, a sentence or two about the distribution in volatility bins would be useful.

Lines 175-177: How many of those species are advected? Increasing the number of advected tracers by 130 and only getting a 16% slowdown of the model sounds too good to be true. Is this 16% degradation of performance a metric for EMAC (the CTM model) or ECHAM-MESSy (the CCM)? What is the total number of advected tracers,

for comparison?

Line 320 and figure 2: Boreal forest emissions maximize during summer. In addition, although the net photochemical activity is probably less in absolute values there than in tropical forests, the length of the day is much longer, and the net photochemical activity there is not negligible. I would expect to see more OA over boreal forests, which are not present there. On the other hand, there are local maxima over very cold and remote regions with high altitude (Greenland, Antarctica) which are probably unrealistic. Is this a combined effect of long-range transport and temperature dependence?

Line 359: Please soften this statement. The way it is currently written implies that this is the one and only reason of underestimation.

Line 413-415: Why OA are less oxidized in the tropics, where photochemical activity (thus aging) is higher?

Line 449: I find the statement that POA is simulated well overly optimistic. The plots in figure 8 don't resemble a straight line, and roughly a third of the data points are under- or over-estimating measurements by a factor of 3 or more. In addition, the data points are not representative for the global atmosphere, since major OA-rich regions are absent, due to lack of measurements there. I have no doubt that the authors did their best to compare the model with whatever relevant data is available, but the fact that the mean bias is small does not mean that very large differences exist when comparing individual stations.

Section 5.2: This is one of the most innovative parts of the manuscript, but stays in just describing the results. Expanding this discussion in implications would be valuable here. Examples include, but are not limited to, why the model performs better during winter, when the OA performance is the worst? What does that tell us for the missing source of OA? Also, how does the O:C ratio compare against OM/OC values used by other models? How can the improved O:C simulation with ORACLES inform other models without VBS capabilities to improve their simulations by probably using different

OM/OC ratios?

Line 501: I do not understand why caution the reader here that there are only 4 field campaigns during summer months, when there are also only 4 during winter and just 2 during autumn (Table 7).

The summary lacks two important conclusions from this work: one, the fact that the model, same as probably all other global models, underestimates OA during winter, and two, the seasonality of O:C.

Code data and availability, line 565: Although it is clear what steps need to be taken if a user wants to use ORACLES in EMAC, there is no mention on whether the ORACLES model is modular enough to be ported in other models, and whether its code is even available for a user (a user is allowed) to do such a thing.

A zonal mean plot for total OA would be a great supplement in Figure 2 and the discussion.

Figure 5b: Why aged SOA maximizes over India?

Technical comments

Please explain what aSOA-v, b-SOAv, SOAsv and SOAiv mean in section 3.4.

Line 241: I believe the correct reference for the OH reaction rate is Donahue et al. (2006).

Section 5 introduction: These equations are textbook material, they should be moved in the supplement.

The second paragraph of section 5.2 sounds a lot like a repetition of the first one, with the addition of some numbers. Please consider revising the text.

Figure 1: What happens with the empty bins, e.g. high volatility and high O:C? Are they being considered in the model, or there is a smart way implemented in the model

to ignore them, which saves CPU time?

Figure 5: It appears some values in the remote oceans have been filtered out. What is the criterion for this? I don't think that there are numerical instabilities in the ratios as presented in the figure for when the concetrations are really low, so I don't understand why filter them out. If e.g. everything is red in Figure 5d over the south Pacific, it is a perfectly valid and expected result.

Figure 8: the 1:2 and 2:1 lines don't meet the axes at the relevant tick marks. Maybe a plotting problem?