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Comment on gmd-2022-90

Anonymous Referee #2

Referee comment on "A lumped species approach for the simulation of secondary organic aerosol production from intermediate-volatility organic compounds (IVOCs): application to road transport in PMCAMx-iv (v1.0)" by Stella E. I. Manavi and Spyros N. Pandis, Geosci. Model Dev. Discuss., <https://doi.org/10.5194/gmd-2022-90-RC2>, 2022

Paper review :

The paper by Manavi and Pandis presents an interesting framework for a lumped oxidation scheme of IVOC (intermediate volatility organic compounds) compounds and subsequent SOA formation. The scheme relies on a review of recent laboratory studies giving SOA yields for oxidation of parent IVOCs. Such a scheme could be implemented into CTMs for better predicting SOA build-up from IVOC emissions. It shows that IVOC emissions lead to much higher SOA yields as from the highest class of alkane and aromatic compounds in the SPARC chemical mechanism. IVOC emissions from the road sector are also evaluated over Europe. The paper should be of interest for the GMD readership and is recommended for publication after several issues have been addressed.

General comments :

The paper is somewhat inbetween a presentation of a general IVOC oxidation framework and an implementation into PMCAMx, for the case of traffic emissions. As such a case study is reserved for a follow-up paper (this seems acceptable), I suggest to present the development as a general one, being applicable to CTM's in general. Then there is no need to explore the case study and May 2008 European IVOC emissions. For instance the emission section 2.5 is clearly engaged in explaining how emission data for the European wide case study are built (however lacking details, see my remark below). I would take a step back, and focus on what is provided by the new IVOC module, how these data can be specific for US, as you already say, and what data is needed to couple this with classical emission data.

My other concern is the benchmark against which the new scheme is evaluated. The proposed benchmark here for yields are the SPARC ALK5 and ARO2 classes for heaviest or heavier alkane and aromatic VOC's. Doesn't this approach mean that you compare to the new model formulation to a model without implementation of IVOC related SOA formation. I would have expected an evaluation against the Robinson et al. (2007) VBS IVOC scheme, which albeit simple, has been probably implemented in many CTMs using the VBS approach. Sure, reference to this scheme is made a many places in the paper, but a comprehensive and quantitative evaluation of differences is not made, and would be an interesting add-up of the study. This could be done in a 0D mode, for exemple, but not necessarily, starting from average European May 2008 conditions. This would allow discussing how the interplay between higher IVOC emissions, lower kOH rate constants and higher SOA yields in the new scheme presented here affects SOA yields with respect to the former Robinson scheme and also to a scheme without IVOC emissions.

Another fundamental difference between the ancient Robinson and the new scheme is that the Robinson scheme only moves mass to next lower volatility class while in the new scheme products can have a large range of volatility. A question : once these IVOC oxidation products formed , do they still further age and subsequently pass to lower volatility classes, as in the initial Robinson scheme ?

Following the philosophy of a new module that could be implemented in diverse CTM models, it would be interesting add a short section indicating the needed model structure and input.

Specific comments :

The paper's title is « A lumped species approach for the simulation of secondary organic aerosol production from intermediate volatility organic compounds (IVOCs): Application to road transport in PMCAMx-iv (v1.0) ». From this I would expect that the new scheme is run and evaluated at least for a case study, but the paper is restricted to model formulation (which is OK). The scheme can also be implemented to other CTM's quite directly as long as they use the SPARC chemical mechanism. I would suggest to make

appear this more general aspect in the paper's title.

Page 6, line 178 :

The volatile products of the reactions of the four new lumped alkane species are assumed as a zeroth approximation to be the same as the ones produced by the reaction of ALK5

Even if this may underestimate mass of gaseous products, still this allows to stick as close as possible to existing gas phase chemistry (still some differences due to different OH reactivities). May be worthwhile to say.

Page 8, line 215 :

the following objective function Q :

$$Q = \sum_i \left[\frac{(Y_{i,meas} - Y_{i,pred})^2}{Y_{i,meas}^2} + \lambda \sum_j a_j^2 \right]$$

where $Y_{i,meas}$ are the measured aerosol SOA yields and $Y_{i,pred}$ is the corresponding predicted yield for the choices of the parameters, using the VBS framework. The objective function Q is minimized by using the *fmincon* MATLAB function (MathWorks, 2020). By minimizing the objective function, the optimal ΔH and a_i 's are determined for the chosen C_i * basis set. »

How accurate is this method ? The method ideally requires that laboratory results cover the range of OA concentrations from 0.1 $\mu\text{g}/\text{m}^3$ to 1000 $\mu\text{g}/\text{m}^3$. For some of the compounds in figure S1, laboratory studies do not cover atmospheric relevant conditions with low enough OA concentrations. Such problems also might have appeared for former studies in the VBS framework, but still the authors should please comment to this question and put some sentences about the limitations of the method.

Page 8, line 230 :

« For the individual compounds lumped in ALK7, ALK8, PAH2 and ARO3 there were no experimental data. »

I think it should be ALK8 and ALK9 instead of ALK7 and ALK8. A few lines above you say that there are data for ALK7.

Page 9, line 249 :

« For example, the mass-based yields of 2,6,10-trimethyltridecane are assumed to be the same as these of n-tridecane. This provides a lower bound for our estimations, as it has been suggested that the SOA yields decrease as the number of branching methyl groups increase ... »

Shouldn't it be a higher bound, following your argumentation ?

Page 9, section 2.5 :

In this section, authors describe IVOC emission factors for specific US conditions, which may be a necessary assumption. In Figure 1, they give spatialised emissions distributions for some compounds and for a given month. Authors should indicate the sources of data needed for such estimations (estimations of total VOC and traffic emissions, vehicle fleet partition for the different classes in Table S1, etc.). The simple reference to the former GEMS project is not sufficient. From table S1, it appears that finally only gasoline and diesel cars are distinguished neglecting differences between passenger and light, medium and heavy duty cars. It seems that bulk European fraction of diesel cars is used, but these fractions are different from country to country. May be at the end giving spatialised emission estimates goes to far given the limited data for this study, and this can be left for the follow-up 3D study.