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Geoscientific Model Development Discussions

Interactive comment on "CAPRAM reduction towards an operational multiphase halogen and DMS chemistry treatment in the chemistry transport model COSMO-MUSCAT(5.04e)" *by* Erik H. Hoffmann et al.

Anonymous Referee #2

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The authors have developed a reduced version of two chemical mechanisms that consider multiphase reactions of dimethylsulfide and reactive halogen species by identifying the primary pathways through which key atmospheric products are formed. The goal of this work is to be able to account for the combined effects of these important multiphase mechanisms in large-scale models for which more comprehensive mechanisms are computationally infeasible. The reduced mechanism is evaluated against a detailed scheme including the two "pre-reduction" versions of the mechanisms under several atmospherically relevant sets of conditions. The authors use the reduced

mechanism to draw conclusions on the various factors contributing to DMS oxidation in cloudy environments and reveal strong direct and indirect effects of clouds on this process.

The work is novel in that it provides a new means to account for the combined effects of multiphase dimetylsulfide oxidation and reactive halogen chemistry in large-scale models. The methodology by which the reduction is performed is well reasoned and follows established approaches for such reductions. The evaluation of the reduced mechanism is convincing and the conclusions drawn from the results of incorporating these chemical processes in a chemical transport model are well argued. It is recommended that this article be published in GMD after consideration of a few comments.

The only major comment is that the grammar in certain sections of the manuscript makes interpretation of the arguments difficult at times. I would recommend that the manuscript be edited for grammar by a native English speaker, or someone similarly fluent in English. Some examples (but not all cases) of such passages are included the following comments.

Lines 80–81: "not only the solvation of the high CPU consumption is necessary"

"Solvation" refers to solvent–solute interactions. Also, it is not entirely clear what is meant here. Is not the point of reduced mechanisms to reduce CPU consumption? Does this refer to efforts to develop more efficient numerical solvers for chemical systems?

Line 83–84: "An adequate mechanism does not currently exist and can only be derived by reducing detailed multiphase chemistry mechanisms."

I'm not sure that I agree that this is the "only" means to generate such a mechanism. For example, have there not been some machine learning-based techniques applied to simulating atmospheric chemical species transformations based purely on observations? It seems these types of approaches could lead to similarly predictive models,

but may shed less light on the underlying chemistry.

Line 91 “the various effects of clouds essentially on halogens and DMS.”

It is not clear what the word ‘essentially’ means here.

Lines 103–104: “The goal of reducing the CAPRAM-DM1.0 and CAPRAM-HM3.0 is that both modules can be applied in different marine atmospheric environments in CTMs.”

This is somewhat unclear. It seems that the results of this work are a single combined reduced mechanism. However, this sentence makes it seem like the goal is to develop two separate reduced mechanisms, and that a CTM can choose which to apply (possibly together or separately?) to particular grid cells. If this is the case, maybe this could also be included in the introduction when the goals of the work are first stated.

Lines 132–140

I find it somewhat unclear how the ‘importance’ of a chemical species is determined here and would prefer a slightly more specific description of why certain species are included in the evaluation. Do you expect that the choice of which species to include in the evaluation, and at what acceptable level of accuracy, would have a large effect on which chemical pathways end up in the reduced mechanism?

Line 268: “As already modelled in other studies, the analyses revealed that the Cl atom is an important oxidant for VOCs and OVOCs”

References for these other studies should be included.

Lines 274–277: “Therefore, a first screening on treated VOCs and OVOCs in the mechanisms MOZART4.0 (Schultz et al., 2018), 275 RACM2 (Goliff et al., 2013), MECCA (JoÌĹckel et al., 2016), GEOS-Chem (Wang et al., 2019),and SAPRC11 (Yan et al., 2019) has been performed for the main VOCs and OVOCs. As a result, only the Cl atom oxidation of the lumped VOCs and OVOCs that are treated within all of these

mechanisms is considered further.”

This could be clarified. Do you first determine which organic species are included in the various lumped species in each model? Do the lumped species in the various models comprise the same sets of actual organic species? How do you determine the rate at which Cl reacts with a lumped species? If one mechanism excludes a specific organic species, are its reactions with Cl automatically excluded from the reduced mechanism? I am not clear on why this is necessary.

Line 317–319: “The evaluation simulations are carried out for 45° latitude with a relative humidity of 70 % under pristine ocean (Hoffmann et al., 2016) and polluted coastal conditions (Hoffmann et al., 2019a).”

In the previous section, several latitudes and relative humidities are modeled. Why are these not used in the evaluation of the reduced model? Can you provide an argument that these scenarios are sufficient to evaluate the reduced mechanism?

Line 378–379: “Consequently, the high CPU time required overlay the CPU time consumption from the reduction.”

This should be clarified.

Line 560 Figure 8

These is an unusually square feature in Fig. 8b for aqueous-phase DMSO directly under the cloud. Is this a result of the way cloud grid-cells and aerosol grid-cells are treated in the model (Fig. 3)?

Line 617–618: “Due to the advection of the stable MS- to the right-hand side of the model domain, the spatial DMSO profile is not modelled.”

Is this not shown in Figure 8?

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