

Reply to comments of Anonymous Referee #2

The paper "Oxidation of low-molecular weight organic compounds in cloud droplets: development of the JAMOC chemical mechanism in CAABA/MECCA (version4.5.0gmd)" by Rosanka et al. presents JAMOC, a cloud chemical mechanism. The paper fits within the scope of GMD and I recommend publication once the authors address a couple of points and provide additional information.

Thank you very much for the helpful comments and seeing the value of our work to the community. Please find in black the original comments and in red our replies.

General Comments

I have a little difficulty in understanding how JAMOC fits within the whole MECCA/CAABA/MESSy/EMAC ecosystem and while I am sure this has been described previously in the corresponding papers, I think it should be mentioned repeated here. A few points come to mind:

1. As I understand it, the JAMOC module is part of MECCA and MECCA itself is a module that can be used in the CAABA box-model and/or in the EMAC global model. In this case, it is perhaps better to remove CAABA from the title (and elsewhere in the text)?

The reviewer correctly states that MECCA chemistry can be used inside the CAABA box model as well as inside a larger 3D model, e.g., EMAC. In this model description paper, however, we only evaluate MECCA chemistry in the CAABA box model. Therefore, we would like to keep the title as it is. The global impact of the newly implemented OVOC chemistry is analyzed with EMAC in a companion paper (Rosanka et al., 2020).

2. If I am not mistaken, MECCA already includes an aqueous-phase chemical mechanism. Is JAMOC an upgrade/extension to it or is it supposed to replace it or run alongside it? This relationship should be clarified, and it should be explained, for example, why it is necessary to create a new module and not simply add reactions to the preexisting aqueous-phase module.

We fully agree with the reviewer that the relationship between the different model parts needs to be explained better. JAMOC is an addition to the aqueous-phase chemical mechanism, not a replacement. The main reason for the confusion is probably the modularity and flexibility of the MECCA system. Model users select an individual subset created from different components of the chemical mechanism. Thus, the new aqueous-phase OVOC chemistry from JAMOC can be switched on or off in MECCA as desired. This is now visualized in the new Figure 3 in the User Manual (included in the supplement). In the revised manuscript, it is now elaborated that JAMOC is considered an addition to MECCA's existing aqueous-phase chemical mechanism.

3. On page 3 the authors say: "The inorganic chemistry for the proposed mechanism is very similar to the inorganic chemistry of the standard aqueous-phase mechanism used in EMAC (Tost et al., 2007; Jöckel et al., 2016) and reactions included in MECCA (e.g. Fenton chemistry), which are not yet implemented in EMAC." This is a bit confusing, as it implies that the reactions in JAMOC are already in MECCA and some, but not all, are already in EMAC. However it does not say which ones, so it is difficult to understand how much of a change has been made. This also suggests that JAMOC is meant to replace the current aqueous-phase mechanism in MECCA and/or EMAC. As per my previous point, the relationship between the different parts of the system needs to be clarified.

The discrepancy can be explained by the different release cycles of CAABA and EMAC. We plan to release the CAABA/MECCA box model version 4.5.0 together with this GMD paper. For our global model studies, the new chemistry was inserted into the current EMAC version 2.54. It will be available to the global modelling community when EMAC 2.55 is released, most likely in early 2021. For clarification, we removed "and reactions included in MECCA (e.g. Fenton chemistry), which are not yet implemented in EMAC" from the manuscript.

The other major comment I have is about CLEPS. The authors describe JAMOC as derived from CLEPS but they don't say how this was done. Were the reactions "hand-picked" from CLEPS (if so on which basis?) or was some reduction procedure applied? It is important that the process is described and the rationale behind certain choices is explained.

We fully agree with the reviewer that this is an important part, which is currently missing in the description of JAMOC. Therefore, we majorly revised the introduction to Sect. 2. The new version now includes a clear description of the reduction principles applied and their reasoning.

I also think it would be more useful and, more informative perhaps, to compare the output of "CAABA with JAMOC" to the output of CLEPS, rather than to the output of "CAABA without JAMOC" (Figure 3 and related discussion). This would allow a better understanding of the accuracy of the reduction procedure, and how much information (if any) is lost when the more explicit mechanism CLEPS is condensed into the smaller mechanism JAMOC.

We agree that a box model study comparison between CLEPS in its original form and JAMOC would be ideal. However, we think that a direct comparison is not feasible due to the following two issues. Even though JAMOC is derived from CLEPS, we additionally expand it to include e.g. the oligomerisation of glyoxal and the gas-phase oxidation of gem-diols. Secondly, in its original version, CLEPS is coupled to the gas-phase mechanism MCM, whereas JAMOC is coupled to MOM. Thus, a direct comparison would be highly influenced by the different gas-phase chemistry (a short comparison between MCM and MOM is presented in Sander et al., 2019).

Minor Comments

line 48: "only a selection of species containing up to four carbon atoms react within the aqueous-phase". Can you explain why this choice was made? I understand one of the reasons is to keep the size of the mechanism relatively small, but why only up to C4 species react and up to C10 species undergo phase transfer? Is the reaction of molecules with high carbon number too slow to matter? Do you actually need to transfer C5-C10 into the aqueous-phase if they don't react and you need to keep the mechanism small?

In CLEPS, on which JAMOC is based, only the oxidation of species containing up to four carbon atoms is represented. In the revised manuscript, we added an elaborate explanation on the selection of species taken into account (see earlier reply to a comment of the same reviewer). In the future, we plan to expand JAMOC to also include the oxidation of species with more than four carbon atoms. The uptake of species containing more than four carbon atoms is still needed to represent their removal by wet deposition in the global model EMAC (for which JAMOC is developed).

line 71: can you clarify the difference between apparent and intrinsic Henry's law constant?

We used the term "apparent Henry's law constant" as a synonym for "effective Henry's law constant". As the latter term seems to be more commonly used, we have changed our text accordingly.

line 86: "Pseudo-first order rate constants for the hydration and dehydration are mainly obtained from the literature". This implies that some were obtained or estimated in another way, please clarify.

For some species, the pseudo-first order rate constants are not known from literature. In these cases, the rates are assumed to be the same as for similar species. In the revised manuscript, this now reads as: "Pseudo-first order rate constants for the hydration and dehydration are mainly obtained from the literature (e.g. Doussin and Monod, 2013). In the case of formyldioxidanyl and hydroperoxyacetaldehyde, the pseudo-first order rate constants are assumed to be the same as for formaldehyde and glycolaldehyde, respectively."

line 141: "In all cases, branching ratios are rescaled to 100%." Can you explain this point better?

From literature, not all products are necessarily known. In order to preserve mass within the model, the branching ratios of the known products are rescaled to 100%. In the revised manuscript, this part now reads: "If products are unknown from literature, branching ratios of the known products are rescaled to 100 % in order to preserve mass."

line 149: how much faster is R11 with respect to R12?

The HO₂ elimination (R11) generally occurs at a rate of about $1 \times 10^6 \text{ s}^{-1}$, whereas the O₂⁻ elimination via OH⁻ (R12) occurs at $4 \times 10^2 \text{ s}^{-1}$. Here, we assume a typical OH⁻ concentration within cloud droplets of $1 \times 10^{-7} \text{ M}$.

line 182: correct "modellens"

Done.

figure 3: the first panel should be sum of OVOC rather than sum of VOCs

Thank you for spotting this typo. We adjusted it accordingly.

References

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