

Geosci. Model Dev. Discuss., referee comment RC2
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Comment on gmd-2021-388

Anonymous Referee #2

Referee comment on "Sensitivity of tropospheric ozone to halogen chemistry in the chemistry-climate model LMDZ-INCA vNMHC" by Cyril Caram et al., Geosci. Model Dev. Discuss., <https://doi.org/10.5194/gmd-2021-388-RC2>, 2022

The paper in review for GMD by Caram and co-workers is a first attempt to include tropospheric halogen chemistry within the global chemistry-climate model LMDZ-INCA. This is a very interesting and well intended effort. But, the current version and modeling fall short of the high standard of GMD. I agree with reviewer #1 on all points, but come to a different conclusion that if the modeling and the presentation within the paper are improved, this can eventually be published in GMD with major revisions.

The main problem with the paper at present is that there are no new scientific conclusions that can be drawn from this modeling effort. I suppose that GMD papers can be based mainly on descriptions of new model implementations, but in the case that this is acceptable, then the model description has to be significantly improved in its clarity and some errors and omissions must be addressed for this to be acceptable.

Specific major comments are detailed below:

L35 - VLSL are not the only source of reactive halogens, this is a mistake. Halogens are also released from sea salt aerosols and iodine is released via ozone deposition to the

ocean surface. These should be discussed and included in the modeling. These are mentioned later (around L135), but this must be clarified here. Polar specific halogen activation mechanisms (snow, blowing snow sea salt aerosols, etc) may not need to be included here for the first global modeling study with LMDZ-INCA, but should at least be mentioned.

L101 - The heterogeneous chemistry on sea salt and sulfate aerosols needs to be more clearly detailed. The basic aerosol chemistry within LMDZ prior to adding this must be described in some detail in order to know if what has been added is reasonable. The aerosol chemistry in the two established global models (CAM-Chem and GEOS-Chem) are significantly different. The method implemented here follows the CAM-Chem methodology. However, this implementation seems to only include recycling on sea salt and sulfate aerosols, rather than actual degassing of HCl and HBr triggered by aerosol acidification (or another proxy for acidity, for example sea salt aerosol age). This approach for reactive halogen cycling is already used in CAM-Chem, but it is not correct compared to what is known to happen in the atmosphere. This is particularly important for chlorine release from sea salt aerosols globally. One early example of modeling HCl emissions from sea salt aerosols is:

Erickson III et al., JGR, 1999:

<https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/98JD01384>

Other processes that liberate photochemically active halogens from sea salt aerosols are also known, see for example the early review of Finlayson-Pitts and Hemminger, J. Phys. Chem. A, 2000:

<https://doi.org/10.1021/jp002968n>

L105: I do not understand the discussion of ice chemistry here, is this ice/mixed phase clouds, snow, or sea ice. What does it mean to ensure bromine recycling in the MBL?

L133: How is HCl emitted from sea salt?

L181 - Section 3.1.1 - This section is poorly written and I cannot tell what is being compared with what. There is not enough information or numbers in this section for this to be considered a chlorine model evaluation. The information in this section should be provided in figures, such as a plot of total Cly in LMDZ-INCA compared with CAM-Chem or GEOS-Chem for the same years/conditions as a function of latitude and altitude.

L201 - Section 3.2.1 - Same comments as for Section 3.1.1. (above).

In addition, the discussion of wanting to capture the impact on ozone rather than the actual chemistry is a puzzling and problematic argument. The focus on ground level ozone, while also discussing the entire tropospheric chemistry cycle for ozone at the same time is also a problem.

L218 - Section 3.1.3 - Same comments as for Section 3.1.1 (above). I don't understand why there is a separate iodine flux section, while the bromine and chlorine fluxes are not discussed. I believe this flux section is actually a chemical budget, but the writing is unclear.

L247 - Industrial emissions of chlorine are known to be important for ground level reactive chlorine cycling (see Thornton et al., 2010, <https://www.nature.com/articles/nature08905>). Why are they left out here?

L252 - I don't understand how the measurements and model are compared in Table S9. Where are the measurement sites, what model grid cells are used? What does it mean

that the measurements are a 24 hour average while the model uses an average over months?

L255 - Table 2 - I understand that the model values are diurnal averages, but more is needed on the measurements (timing of measurement). Are you comparing a monthly average from the observation in some cases with a diurnal average from the model? The wording is unclear and it is possible that the model and observations cannot be compared in this way.

L269 - Section 4 - This section shows why the discussion earlier regarding the main goal of the study is to capture ozone chemistry near the surface is not accurate. In this section the whole troposphere is discussed (however this is not well defined).

L289 - The discussion of the factors that result in different tropospheric ozone burdens from different NMVOC emissions needs to be expanded and discussed in more detail. Is the difference due to NMVOC emissions even larger than due to inclusion of halogen chemistry? If yes, then why were the same NMVOC emissions not used for these runs?

L371 - Section 5 - I do not see any new conclusions compared to past work. This should be improved. The main problems are that (1) the modeling is not advanced compared to past work and (2) there are no new conclusions. Therefore, this paper does not fit the aims of GMD at present. However, maybe the GMD editorial team can agree to accept new descriptions of models without these two criteria being met. In general, it is very good to see more models including halogen chemistry. Therefore, it would be a disappointment if this review process ended with the result that there is no halogen chemistry available in LMDZ-INCA in the future.

Comments regarding citations, co-authorship, and code availability:

- There is a persistent issue of self citations within the paper, while ignoring the wider literature. Examples are on lines 33 and 45, only including self citations for the authors' own groups is not appropriate for these sentences.
- For co-authorship - the description of the author contributions is interesting, but may not rise to the level of the GMD co-authorship guidelines (https://www.geoscientific-model-development.net/policies/obligations_for_authors.html, point 9). Specifically, serving as a scientific advisor for the manuscript may not fit within this description from GMD.
- The code availability on google drive is not standard and this code should be archived on GitHub and Zenodo as noted by the GMD executive editor.

Minor comments:

L19 - halogens should not have an s here

L40-41 - halogens both change the partitioning of NO_x and also react with NO₂ to form species that remove NO_x from the system via deposition or other loss processes. While this is said in the next paragraph, this statement is not quite complete/right here.

L184 - halogens' should be halogen