In Black the reviewer comments and in blue our responses

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 it's clarity and some errors and omissions must be addressed for this to be acceptable.

We apologize for the confusion. We did not state clearly our intent. The aim is to develop a simple and computationally cheap halogen chemistry scheme which is able to reproduce most of the effects of halogen species on climate-relevant species, i.e. ozone, methane via oxidizing capacity, and then explore how halogen chemistry can affect the sensitivity of ozone (and the related oxidizing capacity) to its precursors, a potentially important considerations in chemistry-climate interactions. The challenge is to find the right compromise between the level of realism and details (e.g. species and processes) acceptable for the chemistry-climate applications, and the computing efficiency for the long chemistry-climate simulations. Therefore, we only represent the reactions and species which are dominant for the impact of halogens on global ozone destruction. The impacts of halogen chemistry on key short-lived climate forcers (such as ozone and methane) are still totally ignored in the multi-model chemistry-climate simulations. It is probably time for these models to include some of these effects.

The objective of the paper is to explain the methodology adopted, the potential and limitations of such a simplified model. It is all new and valuable information for chemistry-climate modellers. As such, the paper is well suited for GMD.

Among the list of manuscripts published by GMD are:

- geoscientific model descriptions, from statistical models to box models to GCMs;
- development and technical papers, describing developments such as new parameterizations or technical aspects of running models such as the reproducibility of results;

In addition, we show how iodine, bromine and chlorine chemistry can, beyond their already demonstrated impact on tropospheric ozone, also affect the sensitivity of ozone to its precursors, a potentially important considerations in chemistry-climate interactions. These are novel results.

We recognize that the language was sometimes vague and some formulations were clumsy. We sincerely apologize and thank the reviewer for his/her recommendation. We did a thorough work on the manuscript in order to clarify the intent and better organize it to make the flow more logical. Below we give more details on how we responded more specifically to these general and further specific comments.

Specific major comments are detailed below:

L35 - VSLS 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.

We agree, it has been added to the introduction. Note that iodine release from the ocean is considered via prescribed fields.

L101 - The heterogeneous chemistry on sea salt and sulfate aerosols needs to be more clearly detailed.

The list of reactions has been added to make the paper more descriptive.

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 reacative 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

For this first implementation of halogens chemistry in INCA, we did some trade-offs and incorporated a simplified representation of heterogeneous chemistry on sea-salts and sulfates which are prescribed. We added the chemical scheme to the main manuscript to help the reader to clearly understand what is done and clarified the text. We also clarified in part 2.1.1. that the version of INCA used here does not compute aerosols interactively, they are specified which limits the level of sophistication of the scheme.

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?

We apologize for the lack of clarity here. We mean reaction on ice crystals in clouds. We modified the sentence here and remove the confusing sentence about ensuring recycling in the MBL.

L133: How is HCl emitted from sea salt?

We use the geographical distribution of sea-salt and impose the total HCl emitted to match the one reported by Hossaini et al. 2016.

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.

We rewrote this section to clarify what is compared with what. However, the chlorine chemistry is not central as we first aim to represent correctly the ozone loss, which mainly driven iodine and bromine chemistry. We have thus modified the flow of the paper to make this simplified evaluation of halogens concentrations more logical in the context of the paper and its aim.

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.

We now better explain the strategy in the introduction: what is the most important according to previous studies to represent the effect of halogenated chemistry on global tropospheric ozone and thus better hierarchize the evaluation on elements which are of importance for the ozone budget.

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.

We now try to better guide the reader.

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?

We chose to follow the recommendations of Wang et al. (2019) and not to include anthropogenic emissions in our reference simulation since the total industrial emissions constitute only a very small fraction of the total HCl emitted on a global scale (6700 Gg Cl.yr-1 < 100000 Gg Cl.an-1).

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?

Measurements correspond to average of several 24hours means measured over 1 or 2 months. This is clarified in the legend of the Table (now S3). These results have been aggregated by Wang et al. 2019, we use them in a similar way.

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.

Not applicable, the Table has been removed.

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).

We clarified the aim which is not the surface but the troposphere.

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?

This discussion has been removed as it didn’t bring valuable information here.

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 dissapointment if this review process ended with the result that there is no halogen chemistry available in LMDZ-INCA in the future.

We did not state clearly that the aim was not to improve the pre-existing halogen chemistry schemes or carry out a process study about halogen chemistry. The aim is a simplified and cheap scheme which is able to reproduce most of the effects of halogen species on climate-relevant species, namely ozone, for chemistry-climate applications. This paper effectively does not bring new conclusions regarding detailed halogen processes as the aim is to show that the simple INCA scheme is able to “mimic” the few other complex models regarding the impact of halogens on global ozone. Considering how complex is the implementation of halogen chemistry, we consider it as an important milestone for the model. The results about the impact of halogen chemistry on the sensitivity of ozone to its anthropogenic precursors are also new and relevant for chemistry-climate interactions. It shows that a simplified model can also be valuable depending on the application. We understand from the GMD manuscript type description that such a paper fits within their scope. All this information will certainly encourage other global chemistry-transport and chemistry-climate models to pursue such implementation and explore other chemistry-climate interactions involving halogens. If only halogen schemes that are more advanced (and so more costly computationally) than the few existing ones can be published now, we do not know how the long-standing issue about not accounting the impacts of halogen chemistry on key short-lived climate forcers (such as ozone and methane) in all the multi-model climate studies will be resolved.

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.

We now mention the 3 models having incorporated halogen chemistry and cite more studies done by these models in the introduction.

- 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 description was clumsy, contributions have been clarified and fulfil the GMD standards.

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.

A doi to distribute the code and data will be created if the paper is accepted for publication

Minor comments:

L19 - halogens should not have an s here

done

L40-41 - halogens both change the partitioning of NOx and also react with NO2 to form species that remove NOx 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.

"and also react with NO_2 to form species that remove NOx from the system via deposition or other loss processes" has been added

L184 - halogens' should be halogen

done