The new version of your manuscript is much improved; however the second round of review raised some issues that should be addressed before publication. Please consider the suggestions of reviewer #1, which are worth being tested and could lead to an improvement in your halogen extension. Also, since you stated that improving ozone is the main objective, more effort should be put into the validation part its evaluation.

Dear editor,

The main reproach of the reviewer claiming for major revision is the lack of consideration of a major source of chlorine in the model and the need of a representation, even if coarse, of this source. Actually, this is a misunderstanding since this source is represented even if its dependency to pH is not accounted for. The justification of this choice was detailed in the answer to the first set of reviews and clarified in the text at this time.

The second point, that you underline, is the lack of comparison between model outputs and vertical sonde measurements for ozone concentrations. Here we attempt to include the representation of a complex chemistry, for which some chemical processes, sources and sinks are still under debate in the literature to see how it affects the sensitivity of ozone. The aim of a global chemistry transport model can be to reproduce concentrations but above all their aim is to reproduce the sensitivity of species of interest for air pollution or climate issues to change, for example, in anthropogenic emissions (or climate). A model can have a systematic bias in representing some concentrations but can be fitted for purpose, for example can allow to assess change in composition due to human activity as discussed in the Box 6.1 in the chapter 6 of the WG1 IPCC report (2021). Comparisons between models and observations are necessary and can inform on missing processes, but not everything can be assessed through model-observation comparisons and in particular here by doing comparisons between sonde measurements at specific locations and global model outputs due both to representativity difficulties and systematic biases already identified to represent ozone in extratropical troposphere in the southern hemisphere. A systematic benchmarking of this new versions of the model is beyond the scope of this paper but is systematically realized when such a model takes part to international intermodel exercice.

**Review #1**

This a resubmission of the original manuscript that was assessed by two previous independent referees. There was a general agreement between referees that the heterogeneous component of the model is lacking some key reactions, which are important for recycling and release/re-cycling of halogen species back into the gas-phase of the troposphere allowing photo-chemical destruction of e.g. Ozone ($O3$ ) and methane ($CH4$ ). Thus the accurate simulation of the mixing ratios of Cl, Br and I radicals is a prerequisite towards capturing the effects well with respect to climate impacts. Both of the previous referees suggested a major revision in order to achieve the quality necessary for publication in GMD. Looking at the modifications made to the manuscript I am not sure that issues have been fully addressed from the previous versions. There is the danger of formulating a scheme which doesn't simulate enough halogen radicals which, when applied in e.g. a chemical ensemble to predict CH4 lifetimes, would increase uncertainty in the resulting projections due to the lack of key processes. The authors even admit that the simulated Chlorine component is not optimal, although both Bromine and Iodine have higher chemical destruction fluxes against the main tropospheric reactants therefore the under-estimate in total oxidative capacity is likely low. The evaluation of the scheme against observations is very weak and, this being a GMD development paper, needs addressing before publication to meet the requirements of the journal.

We thank the reviewer for his/her review, please find hereafter our responses. Note that if some modifications asked by previous reviews were not done this is because there was a misunderstanding about what is or not considered in the model and the limitations of the scheme due to these hypotheses (compromise between complexity and CPU+skill of the model). Explanations were given in the reply to the first set of reviews and clarification had been done in the text to address them.
Major comments:

(i) Climate and oxidative capacity is moderated by photolysing light, which itself is attenuated by scattering processes involving clouds and aerosols. A large fraction of CH4 is oxidised in the tropical region, which exhibits lower O3 mixing ratios in the lower atmosphere than the more polluted Northern mid-latitudes meaning that the destruction via halogen radicals becomes most important. Sulphate production in clouds is enhanced by the oxidation in droplets via HOCl(aq) and HOBr(aq), but this process is missing from the update whereas there is a direct link to process important for determining future RF (with the S component in more remote regions being biogenic in origin via DMS oxidation). With low H2O2 in more remote locations, the halogen-sulphate production route could determine AOD over the oceans. Can the authors comment on why this omission was made considering the proposed application of LMDZ-INCA towards chemistry-climate studies.

The aim of this version of the model is to simulate the gaseous chemistry and in particular the oxidizing capacity. Aerosols and in particular the effect of halogenated chemistry on AOD through in-cloud chemistry is beyond the scope of the paper. even if, as underlined by the reviewer, this effect is relevant to determine the total effect of halogenated chemistry on climate forces (and finally on climate). Here, we do not use such a model to compute future RF but Earth System Model. Not all models can include every single process, it depends on the application of the model. As explained in the introduction of the paper, for the moment, none of the models used to assess present and future ozone effect on climate in CMIP includes halogenated chemistry, and very few chemistry-transport models include it to quantify the role  of this chemistry. This work is a first important step before adding more complexity.

(ii) The main acid catalysed routes are missing from the aqueous phase chemical scheme applied in the model, as acknowledged by the authors, but should be accounted for in some way. If the authors do not wish to implement a complex heterogeneous scheme they should apply a first-order generation rate to account for release of precursors e.g. -> $Cl_2$ (g) in $s^{-1}$ (based on available cloud and/or aerosol SAD as a meteorological parameter for parametrizing the extent of activation). This could also be done for BrCl(g) release. This could then act as a guide (and reference) for a scheme applicable in other chemistry-climate models rather than a more simplistic version of a CTM scheme. The influence of applying such an assumption should be shown by comparing two simulations with respect to the tropospheric Cl burden, with and without such reactions active.

The reviewer is right: the acid catalysed route is a major source of reactive chlorine that can not be neglected and has to be considered even if in a very simple way. Actually, we do not neglect this source, it is represented even if it is done in a coarse manner, which is not dependent on the pH. We chose to apply the same methodology as in Hossaini et al . 2016.

(iii) Section 3 : Both the total burdens in Sherwen et al. (2015) and Wang et al. (2021) should be put into the text and tables to show the underestimation in tropospheric Cl burden due to the omission of the heterogeneous chemistry discussed above. This avoids undermining the main conclusion in Wang et al., (2021) in that a complex description of heterogeneous chemistry is necessary for an accurate tropospheric Cl burden. A much more important inclusion would be values from a new run using the assumed release rates of halogen precursors.

The total tropospheric burden is 290Gg of HCl versus 306 Gg in Wang et al. 2021. As far as we can see, the burden found by Sherwen et al. 2015 is not given in their paper. The difference is not due to omission of source but to the fact that we use HCl emission from Hossaini et al. 2015 which is a little lower than the source considered in Wang et al. 2021. This difference is about 5% which is low considering the uncertainty in chlorine budgets.

(iv) Figure 2 shows the latitudinal and zonal mean impact on tropospheric $O_3$ . There is no proof this moves the model towards a better description of the distribution of $O_3$ in the global troposphere. There are $O_3$ profile measurements readily available e.g. at the south pole which could be used to validate the substantial percentage reductions in $O_3$ burden for these latitudes. Currently the reader has no idea as to whether the inclusion of halogen chemistry is needed in this version of LMDZ (which would increase resources needed in any long-term run) or what the potential biases with and without are meaning confidence in the model capturing the correct global $O_3$ distribution is low. I find this not acceptable for a GMD paper, which doesn’t need new scientific findings but rather some evaluation of the update to show the model performs well.

The aim of this work is to better represent the chemistry of the ozone system and its sensitivity to precursors. Chemists know that the halogen chemistry occurs and has a significant impact and is thus relevant to implement in global models. However, global models already underestimate systematically the ozone concentrations in the southern lower troposphere (see Griffiths et al. Figure 3, right panel comparing ozone data and multimodal results in the South Pole and showing a systematic underestimate of about

27%) which implies that the inclusion of halogenated chemistry does not improve the model results. It does not mean that halogenated chemistry does not take place or leads to a worse representation of the chemistry but that neglecting it compensates partly for a systematic problem in global models. The uncertainty in representing ozone by global models in this region exceeds 30% (see Griffith et al. Figure 2 top panel on the right). The aim of a global chemistry transport model can be to reproduce concentrations but, above all, their aim is to reproduce the sensitivity of species of interest for air pollution or climate issues to change, for example, in anthropogenic emissions (or climate). A model can have a systematic bias in representing some concentrations but can be "fitted for purpose", for example can allow to assess change in composition due to human activity for example as discussed in the Box 6.1 in the chapter 6 of the WG1 IPCC report (2021). A systematic benchmarking of this new versions of the model is beyond the scope of this paper but is systematically realized when such a model takes part to international intermodel exercice.

Minor comments (i) When multiple references are used they should always be given in a chronological order throughout the text e.g. ln 28.

The references are given by alphabetical order and then by chronological order for multiple papers with the same first author. The compliance with GMD citations rules will be ensured.

(ii) Most of the reaction rates originate from the standard recommendations apart from some exceptions. For instance, Cl + C2H4 has a IUPAC recommendation but a single study value (which may not be the optimal choice) is applied. Why not use a full set of rate data from the recommendations? The IUPAC reaction for Cl + C2H4 is defined as a third-body reaction, whereas the rate applied here is second-order only. Although the impact of global modelling of O3 will be small, a correct description of rate data should be applied.

We thank the reviewer for this notification. The value will be revised in the next version. As mentioned by the reviewer this reaction is not significant and, actually, is not considered in the other models accounting for the chlorine chemistry.

(iii) There are no references for the assumed uptake parameters given in Table 5, even though many of these parameters have been measured and are available in the recommendations. This introduces a discontinuity in the basis upon which the gas-phase and aerosol-phase chemistry is sourced. The uptake values for various species currently applied are identical for both sea-salt and sulfate aerosol which doesn't seem to be correct. Sea-salt is typically 'wet' considering the Relative Humidity over oceans, with IUPAC providing e.g. an uptake value of 0.6 for HOBr higher than that used here, whereas for 60%wt H2SO4 it reduces to 0.06. Similar differences can be found for other halogen species. This will alter the global chemical budget terms provided.

Despite efforts from experimentalists, there is a lack of experimental data on heterogeneous processing of halogens leading to large uncertainties and a need of hypothesis and arbitrary choices to represent this heterogeneous chemistry (see e.g. Hossaini et al. 2016, Badia et al. 2019). We chose to build on the work of other modelers who gathered experimentally constrained uptake coefficients to build their model or justified hypothesis when data where missing. References for Table 5 are now added.

**Review #2.** The chemical mechanism mainly includes gas-phase chemistry, and thus lacks some heterogeneous chemistry that has been demonstrated to be important for reactive halogen production and loss. Nonetheless, this represents an important step forward for a global chemistry-climate model. Tropospheric reactive halogens have been shown to be an important lever for controlling the oxidation capacity of the atmosphere, so chemistry models should include this chemistry. The chemistry is complex and has large impacts on ozone, making it a somewhat scary endeavor. Thus, I commend the authors for taking this step forward in their model.
They mainly compare their model with other models that have a more complex representation of tropospheric reactive halogen chemistry, CAM-Chem and GEOS-Chem. Despite lacking important heterogeneous chemistry, their model agrees reasonably well with these other models, with some discrepancies that they attempt to explain. Results comparing preindustrial and industrial simulations and the impact of reactive halogen chemistry during these two time periods also is similar to these other models.

My main suggestions are to clarify the chemistry that is included in their model. Specific examples are listed below:

- In Figure 1, does “chlorine contribution to bromine recycling” indicate the three reactions above this statement, or other reactions? If the latter, which reactions are these? Are they in Table 5?

Yes the sentence refers to the 3 reactions (as for the box above, in which “Methane consumption” is the ‘translation’ of the Cl+VOC/CH4)

- Also in Figure 1, “heterogeneous reactions of bromine on sea salt and sulfate aerosols” is stated, but it would be helpful to refer to specific reactions in the tables. I don’t see these reactions in Table 5.

This box refers to the reactions listed in Table 5. The figure 1 is a very simplistic representation of the halogenated chemistry and the main steps to represent in a global model, the aim here is to introduce the paper to non experts but all the details are in the Tables.

- Table 5 lists the reaction probability for heterogeneous reactions. The reaction probability formulation is typically used for first order reactions, but some reactions listed here are second order, so I’m confused. More detail is needed on how the reaction probability is used for these second-order reactions.

Second order reaction rate constants are calculated by assuming that the first reactant is limiting thus, the first-order rate constant is divided by the concentrations of the adsorbed species as commonly done in models (e.g. GEOS-CHEM and WRF-CHEM). We added this information in the paper.

- Sea salt chloride is emitted at HCl. Why is sea salt bromide not emitted as HBr? Sea-salt bromide is the largest primary source of bromine to the troposphere.

Seasalt bromide is emitted mainly as BrCl and $Br_2$, as represented in the model, the emission of HBr from seasalt is minimal, unlike that for HCl (see Saiz-Lopez and von Glasow, Chemical Society Reviews, 2012) In addition, the process is poorly understood, its quantification highly uncertain and some simulations have shown higher discrepancies, in particular in the representation of the vertical gradient of BrO in the tropic probably due to missing processes in the representation of the halogen chemistry (Schmidt et al. 2016). For that reason, this source is not systematically included in model studies (Sherwen et al. 2016, Badia et al. 2019)

Schmidt, J. A., et al. (2016), Modeling the observed tropospheric BrO background: Importance of multiphase chemistry and implications for ozone, OH, and mercury, *J. Geophys. Res. Atmos.*, 121, 11,819– 11,835, doi:10.1002/2015JD024229.

- Figure 5 says that reactive bromine is produced from N2O5, but I don’t see this reaction in Table 5. It is mentioned in the text that N2O5 does not produce ClNO2, but this is inconsistent with Table 5.

Thank you for mentioning that. Actually, N2O5 had been written by mistake on Figure 5 but forgotten from figure 6. It is now fixed. The text was OK explicitly saying that “$ClNO_2$ is emitted by the heterogeneous reaction of $N_2O_5$ on sea salts and produces 2217 Gg Cl.yr$^{-1}$.”