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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Halogen chemistry reduces tropospheric O_3 radiative forcing" *by* T. Sherwen et al.

Anonymous Referee #1

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During the last decade, increasing measurements of concentration and reactivity of halogenated compounds have been made. They have shown that the role of these compounds in the destruction of tropospheric ozone is more important than previously thought. The present paper of Sherwen et al. aims to quantify the radiative forcing of tropospheric ozone by considering, in addition to the chemical "classical" pathways of ozone production and loss, those involving halogenated compounds. This paper is based on a huge and consistent job done to implement the chemistry of halogenated in the GEOS-Chem model as already detailed in Sherwen et al. 2016 a and b. In the present work, the determination of the current and pre-industrial ozone concentration fields is conducted using a 3D model of chemistry transport GEOS-Chem. Whereas it is central for this study, the computation of the radiative forcing seems relatively simple (using a linear relationship between ozone column and radiative forcing) and is just

mentioned in one sentence in the "involvement" part and not detailed in the methodological part. The methodological limits are not raised nor discussed. The authors, considering the ozone destruction due to halogenated compounds found an increase in tropospheric ozone since preindustrial lower than the one obtained when this chemistry is neglected. Consequently forcing of tropospheric ozone is significantly reduced, by about 20%.

The question investigated in the paper is pertinent regarding the field of study. However, several points in the methodology limit the scope of the results. Some key choices are not at all discussed. Furthermore various "shortcuts" in the rhetoric, especially in the introduction shows a misunderstanding of the purpose for which models were originally developed. The discourse justifying this work needs to be reorganized. Finally, the article has no conclusion; appearing incomplete and looking, at this stage, like an extract of publication. If the uncertainties in the current understanding of halogenated chemistry mechanisms are well discussed, it lacks a critical discussion of the other assumptions used in the modelling chain (pre-industrial emissions, calculation of radiative forcing. . .) and a discussion of the magnitude of the results found compared with the range of radiative forcing values given in the IPCC report. In conclusion, the results presented in this study are insufficiently documented and discussed in a critical way to be published in the state. Some items previously mentioned are detailed below.

- The introduction states: "the fact that the models that are used to calculate radiative forcing of tropospheric O3 (RFTO3) do not contain this [halogen] chemistry (Hauglustaine et al., 1994; Levy et al., 1997; Myhre et al., 2013; Young et al., 2013) raises questions over their ability to reproduce tropospheric composition as more and more observations of tropospheric halogens are made". This point is exaggerated because (1) the tropospheric composition is not limited to ozone; (2) The models contain the main sources and sinks of tropospheric ozone (as also shown by your results in Table 2) so they are able to reproduce the main feature of ozone distribution as shown by

comparisons with the observation-based climatology. It does not mean that they do not need improvement like done in the present paper, but such shortcuts undermine the justification; (3) The aim of such models is to implement the state of the art of the chemistry when it is well understood. The models do not and will probably never describe all the atmospheric chemical pathways, but they are useful tools if only considered like that. It is thus unfair to argue that models developed in the 90's were wrong to neglect processes which have been maturely understood recently.

- The radiative forcing is calculated by applying a linear relationship between ozone column and radiative Forcing. This is surprising knowing the vertical gradient of RF sensitivity to ozone. It needs to be discussed.

- The IPCC radiative forcing or ACCMIP ozone column changes should be given with their range of uncertainty. Hence, the radiative forcing found by these authors is within the range indicated by IPCC.

- Much of the uncertainty in the ozone RF comes from the poor knowledge of natural sources (in particular for preindustrial times), we do not know the assumptions considered in this work for these preindustrial emissions. The biogenic emissions, including the crucial soil NOx are not given. The justification for considering that biomass burning is 10% of the current one has to be explained because many recent studies consider rather a 30-50% reduction (van der Werf et al. Climate of the Past 2013, Lamarque et al. ACP 2010) and even, for some of them, higher emissions than the present ones, as in the 'high fire' hypothesis of Murray et al. ACP 2013.

- Too much significant numbers in the RF given in the introduction

- The figures are sometimes difficult to read/interpret due to the color palette (1, 2, 4 and 6).

Sherwen, T., et al. Iodine's impact on tropospheric oxidants: a global model study in GEOS-Chem, Atmos. Chem. Phys., 16, 1161–1186, doi:10.5194/acp-16-1161-2016, 2016a.

Sherwen, T., et al. Global impacts of tropospheric halogens (Cl, Br, I) on oxidants and composition in GEOS-Chem, Atmos. Chem. Phys. Discuss., 2016b.

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