Anonymous Referee #1

Dear reviewer,

We wish to thank you for taking the time to review our paper and provide constructive comments, response to which have greatly helped improve our manuscript.

In the following we present the referee's comments (in bold) and our associated responses to each comment.

- I would suggest the authors provide more information for their simulation results in Section 3. I think at least a map for Iy or IO should be provided in a similar way as Fig.4. Based on that, a detail explanation of how model mechanisms result in the simulated distribution could be given. Such information can largely help the analysis in later sections.

Thank you for your comment. We agree that the figure with vertical distributions and global maps of I_y and IO is worth providing in the paper to support some of the assumptions given in the paper. The figure and description are presented below.
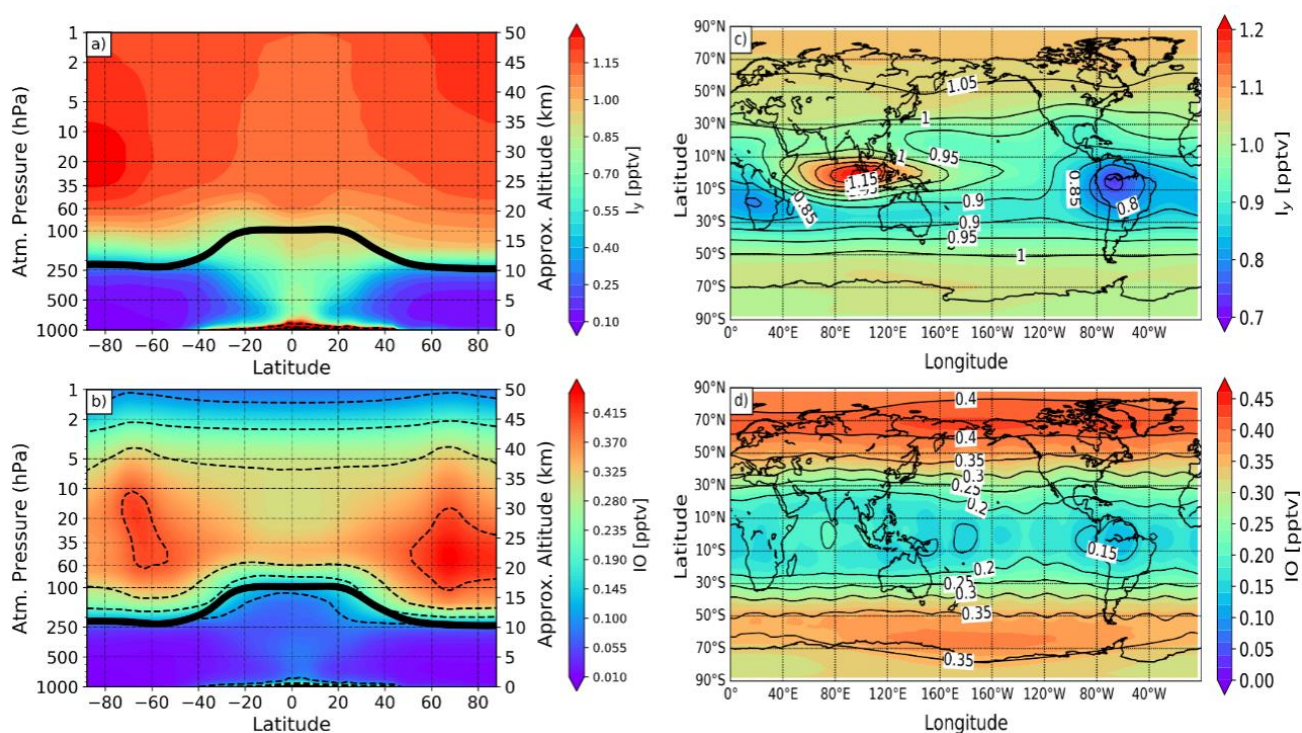

Figure 1. - Modeled vertical distribution and global map of total inorganic gas-phase iodine (I_y) and iodine monoxide (IO) simulated with SOCOL-AERv2-I from the experiment with present-day iodine emissions (1 × iodine), averaged for 2000-2009 period and 10 ensemble members. (a+b) zonal-mean vertical distributions of I_y and IO. (c+d) global maps of I_y and IO averaged for 100-70 hPa region. Black solid line in (a) and (b): annual mean tropopause height.

Figure 1 presents the vertical distributions of I_y and IO at all latitudes and global maps of I_y and IO averaged for the lower stratosphere (100-70 hPa). I_y and IO have the highest mixing ratio at the polar regions of the stratosphere due to transport carried out by Brewer–Dobson circulation (BDC). The larger stratospheric values of IO at high latitudes are also related to the higher O_3 abundance at those locations. At the tropics, there are two pronounced areas with a higher (over Indian ocean) and with a lower (over South America) I_y mixing ratio as depicted in figure 1 c). Their formation might have resulted from different convection patterns (weaker/stronger) over these areas. Also, the area with a higher stratospheric I_y burden is located right over the region with higher surface emissions of HOI/I_2 (see figure 2). In the troposphere, the iodine level is decreasing toward the poles and far from

the iodine source regions. The iodine distribution demonstrates the highest mixing ratio of iodine in the lower stratosphere over middle-to-high latitudes with a maximum I_y of more than 1.15 pptv in the polar region of the Northern hemisphere and about 1 pptv in the Southern hemisphere. The IO has two maximums of about 0.4 pptv in the lower-to-middle stratosphere and high latitudes. Also, note that IO decreases at higher levels of the stratosphere exhibiting more than four times less abundance than in the lower stratosphere, which might result from decreasing the efficiency of O3-I reactions.

- Section 2.2, figures for emission fluxes could be helpful, maybe can be shown in supplements.

Yes, you are right, such a figure will give some insight into the global distribution of iodine source fluxes. We decided to add the figure depicting both organic [CH_3I, CH_2I_2, CH_2ICl, CH_2IBr] and inorganic [HOI/I_2] annual-mean fluxes to the main text (presented below).
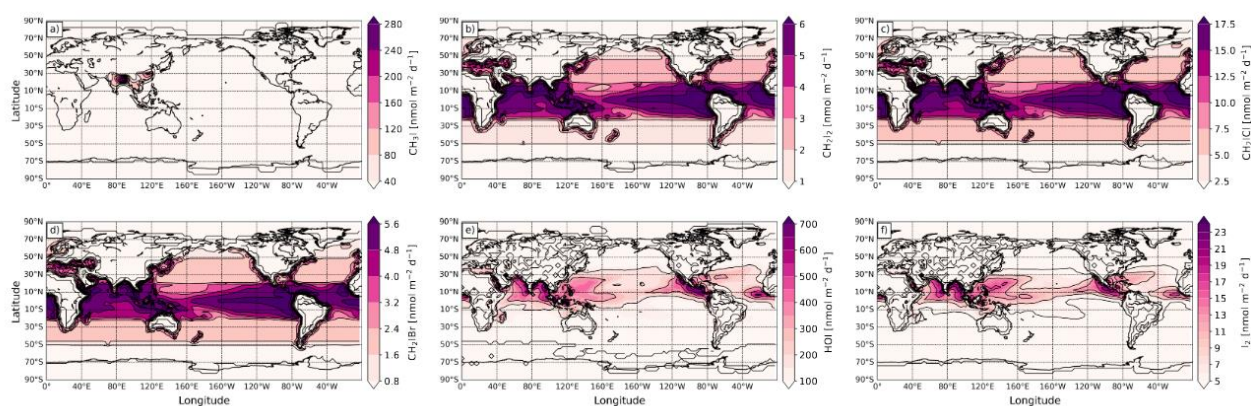

Figure 2 - Annual-mean emission fluxes of organic [CH_3I (a), CH_2I_2 (b), CH_2ICl (c), and CH_2IBr (d)] and inorganic [HOI (e) and I_2 (f)] iodine source gases [in nmol m^{-2} d^{-1}] used in SOCOL-AERv2-I.

- I think the introduction is way too long. As a manuscript for model development, it spends nearly two pages to introduce atmospheric iodine. I would suggest shorten the introduction and refer to the references for the details.

We are not quite sure that the introduction is too long. We think that this kind of paper is worth describing the main up-to-date knowledge and mechanisms related to the behavior of iodine in the atmosphere, including the current state of numerical simulation of iodine to support the importance of this study. However, we agree that some wordings can be changed to decrease the text length. So, we revised the introduction and shortened it by about 15-20% by excluding and reformulating some sentences.

- Line 170, is the 50% underestimation similar everywhere? Will applying the scale factor make the distribution of the emissions different from other models?

We reformulated this description to be clearer. Actually, the SOCOL's ground-level ozone is not uniformly 2 times higher than that from the GEOS-chem model. In Revell et al., (2018), the comparison of ozone from SOCOL to another model and observations is provided. It should be noted that a positive bias of ozone is also exhibited in other models e.g. ACCESS, EMAC-L47, and MRI-ESMr1 (see Revell et al., (2018)). Based on results provided in Revell et al., (2018), we chose to apply the 2 times decrease of the ground-level ozone in SOCOL (that used within the HOI/I_2 parameterization). We still have higher emissions than those in GEOS-chem, but this difference is laid within their uncertainty. It should be mentioned that this scaling factor does not affect the distribution of HOI/I_2 emissions, only the released amount is changed.

Reference:

Revell, L. E., Tummon, F., Stenke, A., Sukhodolov, T., Coulon, A., Rozanov, E., Garny, H., Grewe, V., and Peter, T.: Drivers of the tropospheric ozone budget throughout the 21st century under the medium-high climate scenario RCP 6.0, Atmospheric Chemistry & Physics, 15, 5887–5902, https://doi.org/10.5194/acp-15-5887-2015, 2015.

- Line 241, any reference or potential evidence for this assumption?

Since there is no available information about the deposition rate of iodine species on sulfate aerosols and if consider uptake on sulfate aerosols with the same gamma as for sea-salt aerosols, we would obtain modeled I_y values with a rather large bias (X %) against observations. So, after several model tests, we decided to apply sea-salt gammas divided by 100 to simulate the effective uptake and removal of iodine on sulfate aerosols, which brought the iodine from SOCOL-AERv2-I to be closer to available observations.

- Will the sea salt alkalinity affect the uptake of iodine species?

According to available studies, there is no evidence on the effect of alkalinity on the uptake rates of iodine species as the understanding of uptake rates of iodine species on sea-salt aerosols is still poor (Saiz-Lopez et al. 2012). However, it is well known that heterogenous halogen reactions, including iodine, are impacted by acidity. This will impact the number of source species released (e.g. Br and Cl) or cycled to more labile forms (e.g. I) This is an important point when considering the changes in halogen burdens and impacts over time and the subject of work in the field, for instance, Zhai et al (2021).

Reference:

Saiz-Lopez, A., Plane, J. M. C., Baker, A. R., Carpenter, L. J., Von Glasow, R., Gómez Martín, J. C., McFiggans, G., and Saunders, R. W.: Atmospheric Chemistry of Iodine, Chem. Rev., 112, 1773–1804, https://doi.org/10.1021/cr200029u, 2012.

Zhai, S., Wang, X., McConnell, J. R., Geng, L., Cole-Dai, J., Sigl, M., et al. (2021). Anthropogenic impacts on tropospheric reactive chlorine since the preindustrial. Geophysical Research Letters, 48, e2021GL093808. https://doi.org/10.1029/2021GL093808

- Section 2.2, it seems that the model doesn't include reactive uptake of iodine in liquid clouds. Is this because there is no enough information available for the model parameterization? Maybe make it clearer in the text.

As far as it is currently known, there is no evidence of reactive uptake of iodine species in liquid clouds. The mechanism of heterogeneous reactive uptake of iodine on ice crystals is also still not fully explored and was only proposed as an analogous to that involving chlorine and bromine (Saiz-Lopez et al. 2015). Thus, in our model, we applied the reactive uptake of iodine on ice crystals as was done in Saiz-Lopez et al. (2015).

Reference:

Saiz-Lopez, A., Baidar, S., Cuevas, C. A., Koenig, T. K., Fernandez, R. P., Dix, B., Kinnison, D. E., Lamarque, J. F., Rodriguez-Lloveras, X., Campos, T. L., and Volkamer, R.: Injection of iodine to the stratosphere, grl, 42, 6852–6859, https://doi.org/10.1002/2015GL064796, 2015.

- Figure 1, maybe make the line thicker and the shadings lighter, or use the logscale. It is very hard to see the values in the lower levels.

Yes, thank you, we agree that solid lines in this figure are too thin. We made them thicker. The updated figure is added to the paper (presented below).
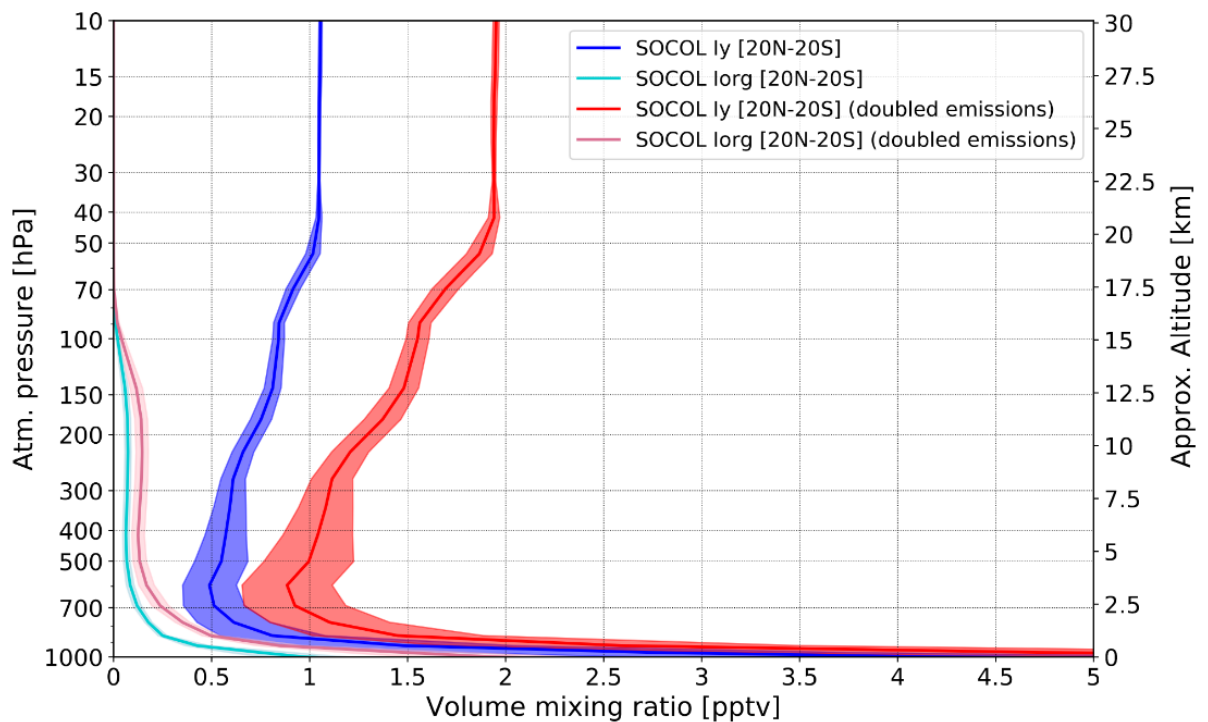
Figure 3. Modeled vertical distribution of total organic (I_{org}) and inorganic (I_y) gas-phase iodine simulated with SOCOL-AERv2-I averaged over tropics [20°N - 20°S], for 2000-2009 period and 10 ensemble members. Red curve: I_y from the experiment 2 x iodine. Blue curve: I_y from the experiment 1 x iodine. Light red and blue curves: I_{org} from 2 x iodine and 1 x iodine experiments, correspondingly. Shadings represent a standard deviation of tropical iodine [20°N - 20°S].

- Line 355-362, this part needs to be explained clearer. The authors referenced a few evidences to show the contribution of cross cycles, but didn't point out whether those evidences were proved in their own model simulation. Maybe provide some numbers to describe the contributions of sole iodine cycles and cross cycles on ozone loss would help.

Thank you for this comment. We agree that the numbers of what is the role of cross-reactions of iodine with bromine and chlorine in the total iodine-induced effect on ozone are worth mentioning here. To do this, we designed the additional model experiment where all cross-reactions of iodine with Br and Cl are excluded from the overall iodine scheme (reaction rate coefficients are set to zero). A % difference in ozone between experiments with/without cross-reactions included related to the no-iodine experiment is presented in figure 4.
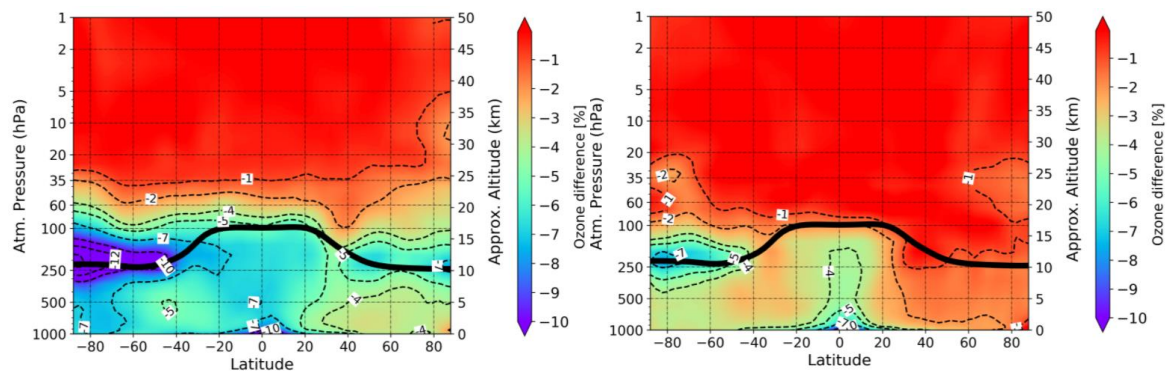

Figure 4 - Modeled effect of iodine chemistry on annual-mean ozone climatology including (left panel) / excluding (right panel) I-Cl and I-Br cross-halogen reactions, averaged for 2000-2009 period. Presented ozone changes of the case with present-day iodine emissions (1 × iodine) relative to the control run (0 × iodine). Black solid line: annual mean tropopause height.

Based on Figure 4, we can conclude that in SOCOL the role of cross-reactions maximizes at the middle and higher latitudes of the lower stratosphere that agrees well with Fernandez et al., (2017) and Barrera et al., (2020). We found that I-Cl and I-Br cross-reactions are responsible for about 60-70% of the total effect of iodine on lower stratospheric ozone in the Northern hemisphere and about 40% in the Southern hemisphere.

Thus, cross-cycles of iodine with Br and Cl themselves are supposed to play an important role in ozone reduction than just chemical reactions of iodine with ozone.

Reference:

Fernandez, R. P., Kinnison, D. E., Lamarque, J.-F., Tilmes, S., and Saiz-Lopez, A.: Impact of biogenic very short-lived bromine on the Antarctic ozone hole during the 21st century, Atmospheric Chemistry & Physics, 17, 1673–1688, https://doi.org/10.5194/acp-17-1673- 2017, 2017.

Barrera A., J., Fernandez, R. P., Iglesias-Suarez, F., Cuevas, C. A., Lamarque, J.-F., and Saiz-Lopez, A.: Seasonal impact of biogenic very short-lived bromocarbons on lowermost stratospheric ozone between 60° N and 60° S during the 21st century, Atmospheric Chemistry & Physics, 20, 8083–8102, https://doi.org/10.5194/acp-20-8083-2020, 2020.

- Line 372, similar as above, the authors referenced Daniel et al. (1999) for the explanation, but did the model simulation in SOCOL show the same?

To demonstrate that the suggestion provided in Daniel et al. (1999) is reasonable for our model results we can refer to Figure 1. In Figure 1, we show the zonal-mean vertical distribution of IO between 1000-1 hPa. This figure demonstrates that the IO mixing ratio sufficiently decreases from the lower to the upper stratosphere. At the upper stratosphere, the mixing ratio of IO is more than four times lower than in the lower stratosphere. This resulted from decreasing the efficiency of catalytic cycles involving iodine. We think that it well supports the argument set out in Daniel et al. (1999).

Reference:

Daniel, J. S., Solomon, S., Portmann, R. W., and Garcia, R. R.: Stratospheric ozone destruction: The importance of bromine relative to chlorine, , 104, 23,871–23,880, https://doi.org/10.1029/1999JD900381, 1999.