Anonymous Referee #2

Dear reviewer,

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

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

This paper describes the implementation of a troposphere-stratosphere iodine scheme in the SOCOL CCM. The scheme is based on the well-used CAM Chem scheme. I think that after the suggested revisions the paper will largely serve the purpose of documenting the model iodine scheme and will be suitable for publication in GMD.

I thought that the evaluation of the model was somewhat superficial and some of the text was not clear. The whole paper would benefit from a thorough proof-reading (see some examples below).

Specific Comments

(1) General. Use of the word 'loss'. The manuscript mentions ozone 'loss' throughout, including the abstract (line 5). Really, what is shown is the ozone difference between a model run with and without iodine. The word loss (like 'depletion') to me implies some time trend or change. (There is also photochemical production/loss rates but that is not what is being shown either). If there was no change in the iodine emissions then this difference would always be present (subject to trends in reaction partners), it was just that models were not so accurate without it. So, I suggest reading through the paper and being clear what is being shown by the difference between the model experiments.

Thank you for your comment and for having read our paper carefully. We accept the point that using the "ozone loss" term without proper context might be unclear while reading the text. So, we edited the paper and reworded it in most of all places to make the text more clear for comprehension by readers.

(2) Line 6-7. Confusing because the number range quoted is globally averaged so we have no idea of the maximising value at high latitude.

Yes, you're right, this sentence was poorly formulated. We corrected this sentence as follows:

"For the present-day atmosphere, the model suggests that the iodine-induced chemistry leads to a 3-4% reduction in the ozone column, which is greatest at high latitudes."

(3) Line 10. Confusing because the sentence appears to be about the lower troposphere but then discusses 50 hPa. Maybe change 'and' to 'but' and explicitly state that 50 hPa is in the stratosphere.

Thank you. Agreed - this sentence is a bit confusing. We revised this sentence and split it into two separate sentences as follows:

"In the lower troposphere, 75% of the modeled ozone reduction originates from inorganic sources of iodine, 25% from organic sources of iodine. At 50 hPa, the results show that the impacts of iodine from both sources are comparable."

(4) The importance of iodine (or not) depends not just on how much ozone might be destroyed by iodine but by any time trend in the abundance. I don't think these results 'constrain' anything – they show the sensitivity.

Thank you. Yes, the word "constrain" might not fit here well. We reformulated this sentence according to your comment as follows:

"Our results demonstrate the sensitivity of atmospheric ozone to iodine chemistry for present and future conditions, but uncertainties remain high due to the paucity of observational data of iodine species."

(5) Line 135. Hadley Centre (spellings)

It was fixed.

(6) Line 149. Use of word 'recur' not clear to me.

In our model, boundary conditions for all organic iodine sources are directly from the GEOS-chem model. They are one-year long and have the monthly-mean temporal resolution. Under the word 'recur' we meant that these fluxes are repeating each model year of simulation. Maybe just the word "repeat" will be more clear?

(7) Line 207. 3 x CFC11

It was corrected.

(8) Line 211. Why 'correspondingly'?

Yes, you are right, maybe the word 'correspondingly' is unnecessary here and can be omitted.

(9) Line 236. Write dt with Delta as in the equation.

'dt' was changed to' Δt'.

(10). Line 253. Experiments. It is commendable to have run 10 ensemble members for each experiment but I cannot see that much use was made of the variability between them. It could be interesting to know how large this variability is. On this point, it is not clear if the SD in e.g. Figure 1 includes this or is just based on the zonal mean of the ensemble mean?

Yes, we agree that the variability between ensemble members of the experiment is missing here. However, the std of iodine between ensemble members of the experiment is found to be less than 1%. We mentioned it in the text but due to it being extremely low it will not be seen in the figure, so we decided not to include it in the line's std. So, we plotted only the std of ensemble-mean iodine between tropical latitudes [20^{o}N - 20^{o}S].

(11) Line 261 'COMPARED to present-day'. Also, why is this a worst case? You cannot assume that. It is just an assumption to investigate the sensitivity.

We use a 2-fold increase of iodine emissions as an assumption for a worst-case scenario compared to the present-day because the prognostic scenarios show less level of future iodine than 2 times of present-day level. At the same time, iodine abundance has tripled over the past 50 years (Cuevas et al. 2018 and Legrand et al. 2018). Nevertheless, we agree with your point that it is a rough estimate and in our study it is only used to assess sensitivity. We also could say that it

is the sensitivity with emissions worse than in the present time but it might not necessarily be "the worst of all".

Reference:

Cuevas, C., Maffezzoli, N., and Corella, J. e. a.: Rapid increase in atmospheric iodine levels in the North Atlantic since the mid-20th century., Nat Commun, 9, 1452, https://doi.org/10.1038/s41467-018-03756-1, 2018.

Legrand, M., McConnell, J. R., Preunkert, S., Arienzo, M., Chellman, N., Gleason, K., Sherwen, T., Evans, M. J., and Carpenter, L. J.: Alpine ice evidence of a three-fold increase in atmospheric iodine deposition since 1950 in Europe due to increasing oceanic emissions, Proceedings of the National Academy of Science, 115, 12 136–12 141, https://doi.org/10.1073/pnas.1809867115, 2018.

(12) Line 264. 'we' -> 'were'?

It was corrected.

(13) Line 276. Figure 1 caption. Last line, why 'ozone'?

Thank you, it was a mistake. Of course, there must be "iodine" there. It was corrected.

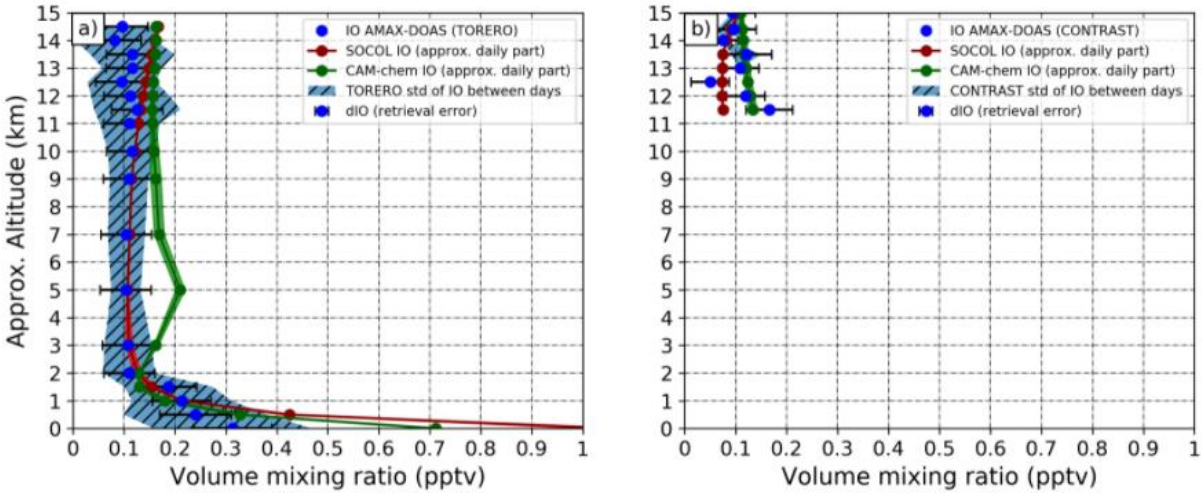
(14) Lines 286 - 291. 'peculiarities' and text around this. This is not clear. The aim of GMD papers is to explain model behaviour and assumptions like this. The text needs clear rewriting to explain if the non-conservation is an issue and at what point it happens or is forced. The blue profile in Figure 1 looks very fixed on 1 pptv. Is that coincidental? The red line seems to show a bit more variation.

Thank you for this comment and for pointing out this issue. We accept that the provided explanation here only reflects our assumptions regarding this issue but do not provide the solid and verified reasons for a gradual increase of iodine in the lower stratosphere seen in SOCOL. We performed several tests to reveal the reason for this. We firstly checked if there is a trend in source gases that might be a reason for the gradual increase seen in the lower stratosphere. Since organic emissions are the same for each year, we checked HOI/I_2 fluxes. The analysis did not reveal the trend. We can also speculate about relaxation time for iodine chemistry but we think that 10 years is enough to reach the equilibrium state. Nevertheless, we checked if there is still an "increase" of I_y burden but comparing the level at 2000-2001 and 2008-2009 period, we found that it is not the case and the abundance of iodine is stable. Also, we tried to use an ideal-age traser to check if there is an issue in the model dynamics. The most possible reason for this increase is the removing of iodine species by interactive wet deposition (by convective cloud's rain) and/or effective/reactive uptake and removing/recycling on ice crystals that are still somehow affecting the transition zone in the lower stratosphere because, horizontally, the removing/recycling is not uniformly distributed since clouds are not everywhere presented and, therefore, stratospheric iodine loading is ubiquitously different. Thus, we could not find the exact process that is responsible for this gradual increase, and in the paper, we addressed only assumptions.

(15) Line 300. The evaluation with the TORERO data is very crude. If there is a reason for this (e.g. free running CCM) then please state it. Why not sample the model like the observations? The 'doubling' assumes the same length of day/night. Roughly ok for the equator but this is just Jan/Feb so will be biased at other latitudes.

Yes, we agree that the comparison against TORERO/CONTRAST observations is crudely performed. To make the evaluation fairer, we limited the comparison to only tropical latitudes [15^{o}N-15^{o}S] as the observations are rather scarce over other latitudes. Also, we sampled model data as observations to conduct an equitable comparison.

The new figure and analysis are addressed in the paper (the updated figure is presented below).



January/February averages of modeled and observed IO in the tropical troposphere [15°N-15°S] for (a) the TORERO campaign from Costa Rica (Jan./Feb. 2012, 10°N-40°S, 250°E-285°E), and (b) the CONTRAST campaign from Guam (Jan./Feb. 2014, 40°N-15°S, 115°E-175°E). Red line: IO from SOCOL-AERv2-I. Green line: IO from CAM-chem. Blue dots: IO observed by AMAX-DOAS. Shadings: IO standard deviations of all modeled/observed IO during the January-February period. Errorbars: AMAX-DOAS retrieval error.

(16). As a general point the model evaluation with observations is very brief. What about data from other sources, e.g. the balloon and ground-based data mentioned in the introduction?

Thank you for this comment. We agree that it would be desirable to add a comparison of iodine modeled by SOCOL with some of the local measurements too. So, we added the comparison of iodine compounds with some of the local measurements mentioned in the introduction as follows:

"The modeled reactive IO over Skandinavia (70°N; 20°E) in March is > 0.45 pptv at 17 km (monthly-mean value) that is in agreement with IO simulated with box model initialized partly with the IO retrieved by balloon flights (a day-time concentration is estimated to be ~ 0.65 pptv at 17 km) despite the measured upper limit of IO mixing ratio of 0.2 pptv (Pundt et al. 1998). SOCOL-AER2-I also captures well the I_y estimated by box model (Pundt et al. 1998) showing a mixing ratio of about 1-1.1 pptv. Also, IO simulated with SOCOL-AERv2-I is corresponding well with DOAS measurements over Spitsbergen island (79°N; 12°E) in March (Wittrock et al. 2000) showing > 0.48 pptv in the lower stratosphere."