Re: acp-2021-876 (The impacts of marine-emitted halogens on OH radical in East Asia during summer).

Dear editors,

We are grateful to the editors and the reviewers for the comments and suggestions. Following is a point-by-point response to the reviewer's comments. Texts in ***Italic Bold*** are the reviewers' comments, and those in normal black are our responses. The blue texts are revised sentences in the revised manuscript. All the line numbers in blue are referred to the change tracking version. We hope that you and the referees will find the changes satisfactory and we are looking forward to hearing from you soon.

Responds to the reviewers' comments

Reviewer #1:

This is the second round of review of the manuscript entitled "The impacts of marine-emitted halogens on OH radicals in East Asia during summer" by Shidong Fan and Ying Li. It is my opinion that the revised manuscript has addressed most of the major issues identified in the original manuscript, and the quality, especially the clarity, of the manuscript is greatly improved. Especially, the major drivers of iodine-induced OH production are clearly identified via Figures 3, 4, and 7, and Figure 8 made some of the key inputs transparent. The authors also carried out additional sensitivity tests (in addition to the nicely designed suite of experiments) to evaluate the impact of anthropogenic emissions on halogen chemistry. I appreciate the efforts the authors have invested in addressing my previous comments. I would recommend this manuscript for publication after very minor (mostly technical) suggestions and clarifications.

1. Line 47 (revised manuscript): "… that when NOx concentration is very low…" Please specify how low is considered as low.

Response:

> "low" generally means that NO concentration below 1 ppbv, but different studies may use different thresholds (e.g., 0.3 ppbv in Tan et al. (2017) and 0.4 ppbv in Rohrer et al. (2014)). We add "(e.g., NO concentration less than several hundred pptv)" in the revised manuscript.

2. Line 72-74: "… the stability of the interaction result of different pathways…" I am not sure what this means. Please clarify or rephrase.

Response:

It means that different emission rates of different halogen species may change the net effect because different pathways are differently influenced by emissions. We change "the stability of the interaction result" to "the net effect".

3. Line 92: Please indicate the pressure altitude of the model top.

Response:

The pressure of the top layer is 50 hPa, which is added in Line 92.

4. Line 116: I could be wrong but I don't think SSA is defined.

Response:

SSA is defined in the Abstract (line 23-24). It is short for sea spray aerosols (just sea salt aerosols in our model). We define it again at the first mention in the main text now.

5. Line 146-148: The authors tested two outdated surface seawater iodide parameterizations (Chance et al. 2014 and McDonald et al. 2014) even though a more advanced data product is available (Sherwen et al. 2019). The authors argued that "Since the reported iodide values by Sherwen et al., (2019) lie between those calculated values according to parameterization of McDonald et al. (2014) and Chance et al. (2014), we chose the latter two to conduct sensitivity simulations." Do keep in mind that the spatial distribution and dynamic range are also very different in Sherwen et al. (2019), especially in the tropical/subtropical western Pacific. I will not ask the authors to perform additional simulations at this stage, but this remains another apparent limitation of this study (both surface seawater parameterizations tested in this work are outdated) that should be noted in Section 3.5 (Limitations of this work).

Response:

Thanks for the comment. We add a paragraph in the revised manuscript to mention this limitation (Line 544-554).

6. Line 149-151: I think Ordóñez et al. (2012) parametrization essentially has constant values in the subtropics and high latitude regions (e.g., north of 20N). I am not sure if simply scaling up based on global emission makes much sense, since much of the discrepancy is driven by spatial variabilities. It would be great to compare the scaled surface seawater halocarbons to the surface seawater observations in this region (e.g., Fuhlbrügge et al., 2016, Fiehn et al., 2017). But I do realize that these halocarbons play a relatively minor role as revealed in this work. Nevertheless, it is worth mentioning this in Section 3.5 (Limitations of this work) that the halocarbons in this region (tropical western Pacific) remains poorly understood which is

potentially important for stratospheric injection.

Response:

Thank you for the great comment. We add a paragraph to discuss the limitations in emissions (Line 544-554).

7. Line 324: "Since these factors are generally species-related…" What factors? Please clarify.

Response:

The factors have been specified in line 310-313 and presented in R1-R4. To make it clearer, we rephrased the sentence from "Since these factors are generally species-related" to "Since these factors just mentioned above are generally species-related".

8. Line 355: Please remind the readers where the Greater Bay Area is. Consider labeling that in the map.

Response:

Thank you for your suggestion. We realize that in a relatively large domain, denoting a small area may distract the attention of readers. Since this information of location is not very important, we delete "the Greater Bay Area" in the revised manuscript and only keep "southern China". Since the Greater Bay Area is in southern China, the statements about maxima or minima still hold.

Rohrer, F., K. D. Lu, A. Hofzumahaus, B. Bohn, T. Brauers, C. C. Chang, H. Fuchs, R. Haseler, F. Holland, M. Hu, K. Kita, Y. Kondo, X. Li, S. R. Lou, A. Oebel, M. Shao, L. M. Zeng, T. Zhu, Y. H. Zhang, and A. Wahner, 2014: Maximum efficiency in the hydroxyl-radical-based self-cleansing of the troposphere. *Nature Geoscience*, **7,** 559-563.

Tan, Z. F., H. Fuchs, K. D. Lu, A. Hofzumahaus, B. Bohn, S. Broch, H. B. Dong, S. Gomm, R. Haseler, L. Y. He, F. Holland, X. Li, Y. Liu, S. H. Lu, F. Rohrer, M. Shao, B. L. Wang, M. Wang, Y. S. Wu, L. M. Zeng, Y. S. Zhang, A. Wahner, and Y. H. Zhang, 2017: Radical chemistry at a rural site (Wangdu) in the North China Plain: observation and model calculations of OH, HO2 and RO2 radicals. *Atmospheric Chemistry and Physics*, **17,** 663-690.

Reviewer #3:

My previous points were mostly adequately addressed and the manuscript was improved. I have the following minor points for further consideration.

1. I previously asked clarification of assumed single scattering albedo used to calculate J(O1D). It seems it was confused with sea spray aerosols (both with acronym of SSA) and thus excluded from clarification. I have tested quick TUV calculations and found that about 5% J(O1D) decrease could occur with Single scattering albedo = 0.95 with AOD = 0.2. I thought the marine aerosols could be brighter (single scattering albedo >0.95). But was that the range that the authors assumed?

Response:

We are sorry for not clarifying single scattering albedo used in CMAQ in our last response. We used CMAQv5.3 to conduct our simulations, which does not need to pre-define single scattering albedo values. Instead, photolysis rates of O_3 and NO_2 are calculated each numerical step. Aerosol concentrations and complex refractive indices of five types of particles (WATER, SOLUTE, DUST, SEASALT, SOOT, see https://www.airqualitymodeling.org/index.php/CMAQv5.1_In-line_Calculation_of_Photolysis_Rates) are used to calculate the optical properties of aerosols. The details can be found in Binkowski et al. (2007) (and references therein).

In old versions of CMAQ (before v5.1), photolysis rates are calculated off-line using a lookup table. Single scattering albedo needs to be provided and it is 0.99 (in the version 5.0.1). Considering the continuity of the development of the model (CMAQ), we therefore infer that the single scattering albedo for sea salt would be larger than 0.95.

2. The explanation about why the InorgI_chem can result in "negative" values over the Philippine Sea was inserted in Lines 481-558 but was a bit too lengthy. Overall, in short, the effect of the O3 level decrease (including in the upstream region) was more important than the influence of additional production of OH from the HOI photolysis?

Response:

In short, yes. In the Philippine Sea, the effect of O_3 decrease is more important than the OH production from HOI cycling and emission. This is also the case in many other oceans because O_3 is generally more “aged” (see e.g., Stone et al. 2018). But in the China seas and Sea of Japan, due to the larger influence of the nearby lands, the effect of O_3 decrease is not fully developed and is therefore less important than the OH production from HOI.

References

Binkowski, F. S., S. Arunachalam, Z. Adelman, and J. P. Pinto, 2007: Examining photolysis rates with a prototype Online photolysis module in CMAQ. *Journal of Applied Meteorology and Climatology*, **46,** 1252-1256.

Stone, D., T. Sherwen, M. J. Evans, S. Vaughan, T. Ingham, L. K. Whalley, P. M. Edwards, K. A. Read, J. D. Lee, S. J. Moller, L. J. Carpenter, A. C. Lewis, and D. E. Heard, 2018: Impacts of bromine and iodine chemistry on tropospheric OH and HO_2: comparing observations with box and global model perspectives. *Atmospheric Chemistry and Physics*, **18,** 3541-3561.