

Review of “Modelling the Impacts of Iodine Chemistry on the Northern Indian Ocean Marine Boundary Layer” by Mahajan et al., ACPD, 2020

The paper presents a computational evaluation of the impact of oceanic iodine emissions (both organic and inorganic) on the composition of the lower atmosphere, with a special focus on the changes on ozone, HOx and NOx (plus NO₃) over the Indian ocean and sub-continent. The most relevant results are: *i*) the requirement to reduce by 40% the strength of the oceanic emissions of inorganic iodine in comparison with previous modeling studies to properly reproduce IO observations performed over the Indian ocean; and *ii*) the different seasonal impact of iodine chemistry predicted during the pre-monsoon and monsoon periods, mostly due to the changes on the total iodine burden during the different seasons. The paper is well organized and the results are presented in order, although in many sections the writing style, figures format and absolute/percentage change description is repetitive, without a comprehensive interpretation on how most of the modeled changes for each individual species are correlated with the others. This work is certainly of interest and fulfills the requirements and objectives of Atmospheric Chemistry and Physics. However, major changes must be performed before acceptance for publication as described below.

Major Comments:

1. Model Validation:

The paper organization and results presentation is correct, starting with the model validation and followed by a model analysis. However, many technical details are missing (both for measurements data and model output processing), which prevents the reader to understand how well the model compares with observations. For example:

- a. How the cruise observations were averaged? Every 4^o Latitdue? Every 2^o Latitude? By looking at Figure 2 (ship-tracks) and Figure 3 (IO and O₃ vmr), it is evident that IIOE-2 and ISOE-8 observations were post-processed in a different way, but no details are given. Which is the temporal resolution of the observations, and how many days of measurements were considered?
- b. How the model output was extracted and processed to compare with observations? As can be seen in Fig. 3, only 1 dataset of model output is compared with 2 independent cruises. As IIOE-2 (Arabian sea) and ISOE-8 (bay of Bengal) campaigns were performed in different seas (and many of the results presented in Figs. 4-10 highlight differences among these regions), wouldn't it be better to compare model vs. observations for each cruise independently?
- c. Did you extract only the closest model pixels to the location of the cruise or did you average a larger region surrounding the ship-lane and/or the whole oceanic domain? Did you consider the whole monthly mean model output to compare with observations, or did you use only a small time-window equivalent to the one for observations? In case the former option, how is the model variability between the monthly mean and the observations-window time period?

- d. The paper presents a model analysis for three different seasons (pre-monsoon in April, summer monsoon in July and winter monsoon in January), while model validation is performed only for winter monsoon when IIOE-2 (December) and ISOE-8 (January) cruises took place. Even when this can be inferred from the text, I suggest explicitly pointing at the lacking of a complete seasonal validation of model results within the model validation section as well as in the Figure caption and discussion. Additionally It would be useful to provide a clear statement within the conclusions that the addressed model impacts for the remaining seasons should be taken with caution, as none the ozone nor the IO results were validated during those seasons.

2. Modeled Geographical Distributions and Statistics:

Once the model is validated, a series of Figures, Tables and Values describing the impact of HAL vs. BASE simulation is provided. Even when in general the presentation is consistent, many of the results are repetitive and presented as a consecutive sequence of absolute and percentage impacts, distinguishing between the whole domain and when a landmask is applied, and written using almost the same style. I believe this description can be improved, making the text more easy-going, pointing at the proper table/figure when results are presented, and mostly by describing absolute/relative impacts altogether as well as making inter-connections between the changes on the abundances of the different species analyzed. In particular:

- a. Many paragraphs of sections 3.4 (NO_x), 3.5 (HO_x) and 3.6 (NO₃) start with “The middle panels of Fig. # show the absolute difference ...” and/or “The bottom panels in Fig. # shows the percentage change ...”. Many portions of this sections present a large series of model mean values \pm standard deviation that can be found in Tables 2 and 3, and in some cases no additional interpretation to the number values is provided. Even when this style can be helpful at the beginning of the results description to orientate the reader on how Figures and Tables should be read, as the paper moves forward it would be very useful not to repeat the full list of values on the text, but just use those that are necessary to justify the analysis being described.
- b. Something similar is present when mean results above the MBL are compared with those over the Indian-subcontinent or the whole model domain. Quantitative results are literally copied from the table into the text for all species and simulations, and usually equivalent conclusions are provided. I found this could be largely reduced to avoid unnecessary repetition, and only include this type of explicit analysis when a special issue must be highlighted, leaving for the remaining of the text a more general comment an inter-connection to the results for other species.
- c. In P15/L345-346 and Table 3 (as well as in other sections below), the “absolute changes rather than the mean changes” are presented. The exact difference between each of the magnitudes should be clearly described at first usage. My understanding is that the “mean change” is the signed difference (HAL-BASE), while the “absolute change” is the unsigned module-difference |HAL-BASE|. If that is the case, I do not completely understand the rationale for providing the

unsigned difference, as for most of the results presented here using a signed analysis that allows determining a positive or negative deviation (bias) with respect to a base simulation would be sufficient. If the authors are interested in presenting statistical evaluation of the model performance (comparing HAL sensitivity respect to the BASE simulation), other statistical measures such as the Normalized Fractional Error (NME) or root mean squared error (RMSE) could be used (see Willmott, 1981. On the validation of models. Phys. Geogr. 2, 184–194. <https://doi.org/10.1080/02723646.1981.10642213>).

- d. Model results are provided for the mean volume mixing ratio within the lower 10 layers within the MBL (P7L156) instead of the surface mean. Even when I'm fine with using this procedure as it allows addressing a more realistic indication of the atmospheric impacts of introducing the organic and inorganic iodine sources on the model, it would be great if the authors could provide an indication of how variable is the vertical distribution within the MBL. For example, all values in the Tables and text present the standard deviation of the model results, but those values are more representative of the "spatial" averaging than the "vertical" averaging. Authors should decide whereas it is worth including an additional figure showing vertical profile or latitudinal cross-section across each of the cruises, but at least an indication of the magnitude of the vertical changes within the MBL would be useful.

3. Iodine Oceanic Source vs Iodine wet deposition:

The authors attribute the strong seasonal variation only to the seasonal change in iodine sources driven by the cleaner oceanic air (lower ozone) during summer Monsoon (P11L258-P12/L266). However, they do not provide any quantitative estimation of the overall source of HOI and I₂ from the ocean during each season, presenting only changes in the IO vmr within the MBL. Even though large seasonal changes on emissions are expected, the Monsoon drives also large changes in precipitation, which in turn will impact on the washout/wet-scavenging of soluble species (and many iodine species are soluble). The authors should be able to quantify the net change in both the fluxes and also the sinks of iodine for the different seasons to support this analysis, or at least highlight the competition between the two processes in determining the Monsoon influence on the iodine burden over the Indian ocean MBL.

- a. Which specific iodine species suffer washout in the iodine chemical scheme, and how different are the modeled/observed precipitation regimes for the pre-monsoon and Monsoon seasons?
- b. I've been also capable of finding a couple of reference to wet-deposition in the text (P14/L316; P25/L580), but only applies to Ozone and NO_x. Equivalent descriptions relating the impact of wet-scavenging on iodine washout should be provided.
- c. The flux strength in the model was reduced by 40%, but it is not explicitly informed how. Did you compute the flux strength at each model pixel using the Carpenter/MacDonald parameterization and then multiply it by 0.6? Just it? Would it be possible/worth to perform an equivalent simulation maintaining the source flux unaltered and increasing the washout by 40%?

Minor Comments:

P2/L27: It could be useful to provide a range of iodine values in the abstract.

P2/L31: Values provided in the abstract are maximum regional changes. It could be of use to mention in the abstract that mean values across the modeled domain are smaller.

P3/L41: I suggest replacing “implicated in” with “associated to”.

P4/L65: “Until recently, the Indian Ocean was **one of** the most under-sampled regions for iodine species ...”

P6/L113: I was surprised the authors did not mention at the end of the introduction that one of the main outcomes of this work was to adjust the iodine source parameterization to obtain a consistent model-observation validation.

P7/L142: “the drastic differences in air masses over the three seasons”. I’m not sure if drastic is the proper adjective to use here, and it should be explicit mention that differences are on the “transport” of air masses.

P7/L145-149: It should be mentioned at some point that the bromine and chlorine chemistry scheme are identical for all simulations.

P8/L168: “discussed further **in** Section 3.2”.

P8/L182-184: “The model captures well the difference between the IIOE-2 and the ISOE-8 cruises, which started from the west and east coasts of India, respectively”. I do not see such a variation for ozone, mostly considering that Fig. 3 shows only 1 set of results for WRF-Chem output without distinguishing between cruises. Could you please explain? Having said this, are there any other ozone observations available (in addition to IIOE-2 and ISOE-8) to compare with model results for the remaining seasons? If no additional measurements are available, at least a comment on the text would help to support the presented implications for ozone.

P9/L196-197: “iodine chemistry would not have any measurable impact”. Why would not instead of does not? You are pointing out to model results that allow to compute the impact.

P9/L204: “However, despite ~~the~~ being an area of high productivity ...”; and leave “by a factor of 10-20 outside the brackets.

P13/L286: Avoid the excessive usage of “only”

P13/L293: “... rather than the photolysis of organiodides, which are long-lived and hence do not contribute heavily to the MBL”. I understand the authors are comparing the lifetimes of organic iodine species with respect to inorganic iodine species. But note that organiodide species are usually referred in the literature as very short-lived species, to distinguish them from the long-lived CFCs and halons. Thus, I suggest rephrasing the sentences to avoid confusion.

P14/L323: “If only the MBL **is considered**, where elevated concentration of IO are observed, **is considered ...**”

P15/L340: Make sure the minus sign sticks to the number within the same line

P15/L349-351: “The reason for larger absolute differences as compared to mean differences is that there are both increases and decreases seen through the domain, and hence the absolute differences gives us an idea of the total impact of iodine chemistry.”. I do not understand the rationale for this type of analysis. See my major comment N^o2c.

P16/L371-373: “The fact that the absolute change values are close to the mean change values shows that most of the domain sees a destruction in ozone due to the presence of iodine compounds.”. Similar to previous comment, I do not understand the rationale for this type of Analysis. If authors want to highlight that iodine-driven ozone destruction is larger than production, this is already shown in Fig. 5.

P16/L374-382: The different percentage impacts in comparison with other studies is well oriented and highlights the different chemical treatments between studies. However, I suggest including here an explicit mention to the fact that the iodine source parameterization has been reduced here, which clearly affect the percentage impact obtained.

P18/L414-415: Why the NO_x abundance over the shipping lanes are more marked for NO than for NO₂? P18/L418-419: Why January show the lowers NO concentration over the shipping lanes? Correct the typo on P18/L422 to make reference to NO instead of NO₂.

P18/L424-426: The authors attribute the smaller standard deviation over the MBL to the “much cleaner air than above the Indian subcontinent”. Could the smaller deviation also be related to the less pronounced day/night variability of dominant NO_x shipping sources compared to continental NO_x sources?

P19/L441: “Over the MBL too, ...” Please rephrase.

P19/L446-448: However, similar to NO₂, these values are misrepresentative of the effect of IO because of differences in the sign of the change across the domain.”. I do not understand the rationale for this type of analysis. See my major comment N^o2c.

P20/L457 and P23/L536: shift the position of HAL and BASE to make it consistent with the percentage change computation.

P20/L458: “decreases in NO_x as high as 50%...”. Was this value computed for the model monthly mean? If that is the case, which are the maximum differences for a specific day or hour?

P20/L471-473: Using “slightly higher” is confusing, as comparatively, the percentage change in July is more than 5 times larger when only the MBL is considered. Also, note that for all this values, the standard deviation is much larger than the mean, highlighting the huge variability on the averaged values, so interpretation should be taken with caution. This should be highlighted in the text.

P20/L475-479 and P26/L603-605: I would expect that, in addition, differences between Li et al., 2019 and the present work are affected by the larger oceanic fraction of the model domain for the Indian Ocean study in comparison with the mostly continental European domain of Li et al.

P23/L543-545: The way the sentence is written seems to indicate positive differences when they are negative. I suggest rephrasing.

P23/L547-551: "The mean percentage change in the OH and HO₂ mixing ratios peaks at 2.6 % and 8.4 % for the months of April and July, respectively (Table 2), but the absolute percentage change in OH is higher at 3.6 % in January, while the HO₂ absolute percentage change (Table 3) is about ~8.4 % showing the large impact of iodine chemistry on the oxidation capacity of the MBL." I do not understand the analysis and implications. See my major comment N^o2c.

P26/L607: Are 24 hr mean used or only night-time values considered?

P26/L610-612: Why the larger changes in NO₃ are predicted during the period of time when iodine chemistry is less active? Is this a day/night issue?

Figures and Tables:

Fig. 1, 4-10: The indication of longitude in all panels appears in the middle of the domain and not at the axis, which makes it very difficult to read. Also note that all figures captions start with "Model simulations showing ..." and ends with "... are shown". Replace lower panels by bottom panels. Please rephrase.

Fig. 2: It might be possible to include the cruise tracks on any of the panels of Fig. 1 to reduce the number of Figures.

Fig. 3: I get a confusion with the "empty squares" symbols for IO, as they are supposed to be "upper limits" but at 12°N and 8°N there is a whisker-range line also for larger values than the square.