

## ***Interactive comment on “Simulations of anthropogenic bromoform indicate high emissions at the coast of East Asia” by Josefine Maas et al.***

### **Anonymous Referee #1**

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Maas et al. 2020 presents an estimate of near-coastal flux of by-product  $\text{CHBr}_3$  emissions from power plant discharges in Asia and its impact on atmospheric bromine loading. This analysis is based on some recent water sample measurements from power plant cooling water and surrounding waters, with the help of Lagrangian trajectory model calculation. This is an interesting study and provides some helpful information in terms of quantifying the anthropogenic contribution to the atmospheric bromine budget. However, I have several major concerns on the method, lack of adequate comparison for the most important region in this study (East Asia), and the major conclusion. These concerns should be addressed before the paper is considered for publication in ACP.

1. Section 2.3. The authors mention that the FLEXPART is run using the meteorolog-

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ical input stem from the ERA-Interim reanalysis . . . The FLEXPART simulations were performed for the boreal winter and summer seasons, for a total of three months with a one-month spin-up. Credible estimate of contribution of surface to UT/LS transport rely on the use of a model that can properly represent this transport process. At minimum, in the case no transport evaluation is conducted for this study, you need to provide adequate peer-reviewed results showing FLEXPART-based analysis is suitable for this study; that it is adequate in representing the surface to UT/LS transport within the Asian tropical/subtropical deep convection and the Asian summer Monsoon over the continent.

2. Second, please state clearly the year of DJF & JJA months you are using to drive the FLEXPART simulation. I am also not convinced a single year (with only 2-seasons) simulation is statistically adequate to quantify the transport from surface to the UT/LS in Asia. The authors need to decide an appropriate length (number of years) to address such transport using FLEXPART and provide a discussion on the year-to-year variability of the above transport. My suggestion is that at minimum you need a 10-year simulation to cover a few full cycles of QBO and ENSO, which have significant impacts on the dynamical transport relevant to this study.

3. This study is based extrapolating the information from a limited number of power plant effluent to the entire Asia power plants. As discussed in section 5, both the simulated oceanic and marine boundary layer concentrations of CHBr<sub>3</sub> from this study, particularly those from the HIGH scenario, are larger than most of the previous observations in general. The regions that where 90% of the largest simulated concentrations (see Figures 5, 6, 7) display extremely high level of bromoform levels compared to the original Ziska 2013 results. Yet, no comparison with previous measurements were presented in this work. NASA has recently conducted an aircraft field campaign KORUS-AQ (<https://www-air.larc.nasa.gov/missions/korus-aq/index.html>) in this region with extensive airborne measurements of CHBr<sub>3</sub> (from the Whole Air Samplers, PI Donald Blake) from surface to mid/upper troposphere that is highly relevant to this

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study. These measurements are publicly available at <https://www-air.larc.nasa.gov/cgi-bin/ArcView/korusaq#BLAKE.DONALD/>. I strongly encourage the authors use the KORUS-AQ CHBr<sub>3</sub> measurements to evaluate the simulated FLEXPART CHBr<sub>3</sub> from the three scenarios for a proper assessment of the design of this experiment to see whether the extrapolation method used in this work is a reasonable approach.

4. Section 4.2: the discussion on the vertical transport of bromoform in the troposphere. While tropical deep convection plays an important role in vertical lofting near the EQ, vertical lofting in subtropical Asia and East Asia is primarily driven by the Asian Summer Monsoon in the summer season. These transport processes were not discussed adequately in this work and past literature were not referenced either. Please add.

5. Figures 8 and 9 and related discussions. Using a climatological cold point altitude of 17km for discussion of vertical lofting and entrance to LS is not adequate, and this is particularly not suitable for the subtropical box (Figure 8). The tropopause in this region is likely very different from the tropics and can be highly variable due to seasons or other dynamical processes. I would suggest the authors to use the tropopause height and potential temperature fields from ERA-Interim reanalysis. Only when the vertically lofted air mass crosses the tropopause and enters beyond the 370-380K potential temperature, the amount of the remaining CHBr<sub>3</sub> within the air mass would have a chance to survive the transport process, make it to the stratosphere and have an impact on stratospheric bromine loading.

6. Final major comment on the main conclusion of this work. With all the previous potential issues I have noted above, the authors concluded that these anthropogenic emissions only contribute 0.02-0.03 ppt to the stratospheric bromine budget. I find it not convincing, from the results presented in this work, to draw the conclusion that anthropogenic sources are important enough to be considered for future estimates of atmospheric bromine input. While local concentrations are high, due to the lack of efficient vertical delivery mechanism, these emissions have little chance of reaching the

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stratosphere. This has been the conventional understanding on the vertical transport efficiency of very-short-lived bromine species, and is seemingly confirmed again in this study.

Minor comments: 1. Lines 60-62. These statements are missing proper references. 2. Lines 149-152: Please list what are the non-volatile and volatile DBPs considered in this experiment 3. Figure 1. It would be helpful if you can add the locations of Table 1 results (the ones that in the region) on this plot, marked with a different symbol.

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