Review of Mass et al.

This paper describes the estimation of sea surface concentrations of bromoform over East Asia resulting from the treatment of power station coolant water with chlorine-based disinfectants. The authors subsequently then estimate sea to air fluxes and test these within an atmospheric transport model to estimate the transport of bromoform in the atmosphere, in particular, via convection to the stratosphere. The authors estimate the sea surface concentrations using a bottom-up approach by first estimating bromoform within power station coolant waters, the discharge of this coolant water into the ocean, and then its subsequent transport in the ocean using oceanic transport model. Based on the bottom-up approach, the authors find a notable contribution to oceanic bromoform from this source that strongly affects oceanic bromoform concentrations close to the source region. Based on the sea-to-air fluxes and the atmospheric modelling, despite showing significant anthropogenic bromoform levels near the source regions and in the boundary layer, the authors find only a modest contribution of this source to stratospheric bromoform mixing ratios.

Bromoform has an important relevance to stratospheric ozone depletion. This paper therefore covers an important topic since current knowledge of bromoform sources is currently highly uncertain due to only limited monitoring of this gas. Attempts to improve our knowledge of its sources are therefore highly welcome. I therefore find that the paper sits well within the scope of the journal.

The subject matter and overall concept and methodology of the paper alone make it worthy for publication. On the whole, this is a good piece of work. Despite the manuscript's strengths though, there are several weaknesses in the work. I therefore have a list of general and specific comments that will need to be addressed to improve the manuscript to a sufficient level to merit final publication.

General Comments

1. **The overall framing of the paper**. I think it would help ease some concerns (see for instance reviewer #1's comments) if the paper's main findings regarding anthropogenic bromoform were to be framed as a set of predictions (that can be tested) based on independent knowledge and data. Your bottom-up method makes a set of predictions based on the data you have used. Then you have made efforts to validate those predictions using sea surface bromoform concentration and atmospheric measurements; these data provide somewhat limited but promising support for your predictions. As it is, the manuscript abstract and conclusions contains various statements that are rather definitive, e.g., "We find that bromoform…" (line 26in the abstract) and, similarly, "We find that…" (line 388 conclusions). When in fact the observational support appears promising yet far from definitive, and would require more dedicated observational monitoring to really prove this hypothesis. I therefore propose the authors modify the manuscript so it clearly follows this chain of reasoning: prediction/hypothesis (emission atmospheric, oceanic modelling) into observational support, followed by evaluation, and then lastly clear statements on what is needed to provide stronger support for this hypothesis, i.e., more specific and targeted observations.
2. **Treatment/explanations of the UTLS and cold point**. I think that the UTLS and cold point definition is too simplistic. The use of UTLS throughout does not capture any of the essential details of this complex atmospheric region. Sticking to a single altitude of 17 km for the cold point is also not really realistic when looking at different latitudes and seasons. Furthermore, sticking to the cold point as a definition of the lower stratosphere alone is also not entirely suitable and the suggestion from reviewer #1 to simply use the 380-390 K potential temperature line also misses some of the subtle complexity. I recommend that the authors consult Corti et al. 2005 and 2006 (see below) that both provide clear explanations and observational support for more nuanced explanations of dynamical interactions in the UTLS. First, the level of zero radiative heating (LZRH) is also useful a measure for whether air masses will undergo slow radiatively driven ascent to

above the 380-390 K levels, and it is usually at 15 km for clear sky conditions. Second, they show that in-cloud (cirrus) radiative heating can be responsible for lofting cloud containing air masses from as low as 11 km upwards to eventually reach the stratosphere. I would recommend that the authors try to calculate the LZRH using the ECMWF meteorological fields to try to diagnose this to help determine which airmasses at altitudes below 17 km are heading up or down; this would really strengthen the paper and strongly aid the interpretation of the results, which is quite difficult at this point. If this is not possible it would be very useful to see at least the 11 and 15 km levels, but this would be a much weaker alternative.

3. **Many missing details**. There are several missing important details from various sections of the paper, e.g., the year that is studied – this is not mentioned at all. I have addressed each of my concerns in specific comments below.
4. **Clarity of the manuscript**. At many points the information given is insufficient to understand precisely what is being said. I have made various specific remarks below to help address this concern.
5. **Duration of FLEXPART simulations/transport times to the stratosphere**. The duration of the FLEXPART runs is 3 months. As shown in Corti et al. 2005/2006, transport times from the boundary layer to the 390 K level of 50 days, and so even with a 1 month spin-up, we can expect a delay of ~20 days for air masses from the beginning of the spin up to reach this level. Indeed, this appears to be visible in Figs. 8b and 9c and d. This complicates the interpretation of the results both for the 5-day averages (i.e., when are they in the course of the simulations), and for the time series in Figs. 8 and 9. Similarly, the bromoform emitted in the last ~50 days of the 3 month simulations has no chance to reach these altitudes. Please discuss these issues.
6. **Discussion of key results**. Despite being an important component of the bromine lofted to the stratosphere resulting from bromoform, the authors make no mention of product gases (PGs). This is particularly important in light of comment #2 above. I do not expect the authors to simulate PG formation, chemistry, washout, and transport, but it should be clearly explained that we expect much of the bromoform to be chemically processed into PGs during the 50 days or so of vertical ascent to 390 K from the boundary layer. Further on this point, it would be worth having some discussion on CTM studies showing the partitioning of bromoform and PGs at different levels in the atmosphere.
7. **Year of study**. As mentioned, the year of study is not mentioned in the paper. While it could be interesting to do a multi-year analysis, if this is beyond the capabilities/time constraints of the authors, an alternative would be to provide some climatological context on the specific year of study. The WMO annual climate reports usually give a good region by region analysis that would help to set the meteorological context.

References

Corti, T., B. P. Luo T. Peter H. Vömel Q. Fu., Mean radiative energy balance and vertical mass fluxes in the equatorial upper troposphere and lower stratosphere, Geophysical Research Letters, https://doi.org/10.1029/2004GL021889, 32 (6), 2005.

Corti, T., Luo, B. P., Fu, Q., Vömel, H., and Peter, T.: The impact of cirrus clouds on tropical troposphere-to-stratosphere transport, Atmos. Chem. Phys., 6, 2539-2547, https://doi.org/10.5194/acp-6-2539-2006, 2006.

Specific Comments

I found the abstract to unclear and at time contradictory from line 20 onwards. I think this stems from the fact that the authors try to say a bit too much at the same time while also only partially introducing terms such as “bottom-up estimates”. Here, this is a specific reference to the prior work Ziska et al.

2013, but when I initially read this it appears to be a reference to the method in the current work since one could also classify this as a bottom-up inventory set of sea-air fluxes.

I found issue with the single number 0.03 pptv of bromoform in the stratosphere quoted in the abstract. Firstly, given that two scenarios are discussed (LOW and MODERATE), it seemed odd to only quote a single value. Further to this point, the authors look at two different seasons yet only quote one value – again, please resolve this issue. Second, if this is an average, it would make sense to quote the associated standard deviation. Third, there is no context or explanation given for this number: is it a temporal average, a spatial average, what is the duration of the average? These are all important details that would allow readers to understand the results.

Line 43. It would make sense to show some of the chemical equations associated with bromoform formation in coolant water if they are known.

The ordering of the introduction was a bit disjointed in my opinion and to also contain some text that is not relevant to the work at hand. I would remove the sentences between lines 50 and 55. In my opinion the text should be reordered such that the paragraph on lines 70-82 should be the second paragraph. The third paragraph should then be on lines 56-68. This would make a more logical flow in my opinion.

Line 117. It was not clear what you meant by the settlement of pathogens. Do you mean growth?

Line 137. I do not claim that this is important to their findings, but the authors should justify not including diffusion.

Line 140. Please mention the years you are looking at in this study and in Mass et al. 2019.

Line 149. I could not make sense of the following text. It was not clear how point 2 relates to the text that follows or where point 2 is discussed. It was unclear what "distinguish" meant in this context – this is too vague and a more precise explanation would be welcome. Are points 1 and 2 meant to describe separate simulations? Separate processes? And why are 1 and 2 being treated separately at all? Clearer explanations here would be very helpful to the clarity of the manuscript.

Line 171. The authors should make it clearer how the values of C_{eq} are calculated from the outgassed bromoform; this is currently not explained.

Lines 167-173. In general, this section of text needs to be clearer. This could be improved by stating that the low C_{eq} values are driven by low atmospheric vmr. It would also be clearer if the authors stated how C_{eq} relates to vmr.

Line 178. "Mean concentrations are calculated…". In air, C_W, or C_{eq} or atmospheric vmr?

Line 178-179. "…of bromoform, characterised by the highest local concentrations, accumulate." This is not very clear.

Line 180. "Mean and maximum fluxes are calculated based on the same principle." What principle?

Line 181. "The annual mean atmospheric bromine input from industrial bromoform emissions". I think you mean *resulting* instead of "input".

Section2.3. We are missing a lot of details here. What resolution are the simulations carried out on? The same resolution as the meteorology? Are the emissions constant during a season? Are Lagrangian particles emitted over the entire ocean and then the emission rate is proportional to the air-sea flux? What year are you looking at?

Line 195 onwards. We are told that there are three additional runs that are made. Then, over the course of four paragraphs with at times unclear descriptions we are told details about them, but they are only referred to as 'first run' and then 'two additional runs', and then 'first of two runs'. These descriptions are imprecise and confusing. Please can the authors define three names for the runs in

line 195 first and then describe them in the following text as “Run A does this….Run B does that … etc”.

Line 214. The authors refer to means of the whole domain, but what is the domain?

Line 214-215. I could not understand the descriptions as they are for “Mean mixing ratios from the whole domain in the marine boundary layer and in the UTLS are given as the average over the 90 % area characterised by the highest local values, and maximum mixing ratios as the average over the largest 10 % (see Section 2.2).” Also, how did the authors decide upon the 90% and 10% levels?

Line 216. The authors say they identify two regions. I think they mean *define*.

Line 221. “…pattern in the research area of interest (Figure 3).” I think the authors mean region, and also which region? There are different areas being talked about. Please be precise for clarity.

Line 231. Please can the authors show the Kuroshio current on the map?

Lines 259-264. The section is unclear. The sentence on lines 262-264 is particularly unclear. Also, for clarity sake, please refer consistently to the Ziska et al. 2013 emissions as Ziska2013. These sentences are confusing because information is expressed imprecisely and there are references to prior statements that themselves unclear. Please try to arrange the information clearly, methodically, and logically.

Line 264. Is the implication of the results that most of the East Asian CHBr3 in Ziska2013 is anthropogenic in origin? I think the authors should state this more clearly if this is the prediction.

Line 266. What is the 29% percentage relative to?

Line 271. I found it odd that the authors make a 3 month long simulation and then only show a 5-day average in that entire simulation. Please can the authors explain or justify why such a short period of time is selected? Could the authors consider either monthly or 3-monthly averages as well? Also, which 5 days is this from within the 3 month simulation? All instances of this should be made clear and/or justified.

Lines 288-299. The authors discuss Figure 6 in relation to this text but do not mention the DJF results in Figure 7.

Lines 302-303. From the description given, it is not entirely clear what has been averaged. I assume it is a spatial average, but the authors should specify because the sentence implies it is spatial and temporal.

Sentence on lines 321-324. I suggest placing this sentence prior to the sentence beginning “Thus, …” on line 320.

Line 333. Please explain when and where the 5-day snapshot is.

Line 340 and 343. Please state when and where the vmr values are calculated for.

Lines 363-369. I am concerned here at the averaging approach reduces the complexity and is masking effects of over sampling of the open ocean regions. Thus, I am not sure this shows a good comparison of the same thing. I think this highlights that more thorough statistical analysis needs to be carried out, i.e., a simple x versus y spatial scatter plot. Including this would strengthen the conclusions of the paper.

Line 378. There is no mention of the year under comparison. Providing that there is overlap in the year, the KORUS-AQ data suggested by reviewer #1 could be useful here.

Line 388. Recommend changing “find” to *predict*.

Line 392. Make sure it is clear these are simulated vmrs.

Line 392. What is a cloud of high bromoform? Perhaps use something more precise like “A diffuse area with high bromoform abundances”.

Line 395-396. Please be more specific as this sentence is unclear.

Line 403. Recommend stating that the assumptions are reasonable in the majority of case since the cited observations show larger ranges than those stated here.

Line 406. Recommend stating that the HIGH results are only too high in the majority of cases.

Line 408. Recommend being more specific. Instead of "results" state *bottom-up emissions, modelling, and observations*.

Technical Comments

Recommendations. Please use a comma after uses of which in cases where it introduces a nonrestrictive phrase. When describing using a method from another publication use *following* instead of after.

Line 10. Modify to "…have increased rapidly exceeding mean global growth."

Line 36. Modify to "Discharge of DBPs within the cooling…"

Line 40. Modify to "…regularly involve the discharge large volumes of water into the marine environment."

Line 41. Modify to "…and its decreased density means it is at the sea surface. Chemicals such as DBPs contained in cooling water are likely to spread laterally…".

Line 83. Modify to "…contributions to VSLSs, in the form of…"

Line 84. Modify to "…50 % of the global coastal cooling…"

Line 87. Modify to "…we show oceanic distributions"

Section 2.1 title. Recommend changing to "Estimation of DBP production in cooling water from East Asian power plants".

Line 96. Modify to "…the ocean provides an unlimited water supply."

Line 136. Modify to "…discharged with the cooling water."

Line 170. Modify to "…the impact that atmospheric bromoform abundances have upon on the flux calculations"

Line 185. Modify to "…bromoform for the three different emission strength scenarios with the Lagrangian…"

Line 187. "…(temperature, and winds)…"

Line 210. "…than the Ziska2013 emissions."

Line 218. "…and over another region from China…"

Line 219. "…we refer to this as the subtropical box…"

Line 221. "…in the region of interest…"

Line 222. "Non-volatile DBPs from cooling water usually accumulates…"

Line 229. "…in the South China Sea suggests only small contributions…"

Line 234. "Figure 3, because the volatile DBPs…"

Line 235. "…for the three emissions scenarios LOW…"

Line 236. "…smaller spread compared to the non-volatile DBPs."

Line 263. "…the Ziska2013 biogenic emissions are spread out…"

Line 264. "…similar total emissions as in the LOW emission…"

Line 283. "…the three scenarios…"

Line 292. "These differences are maximised…"

Line 319. “…in the tropical marine boundary layer where mixing ratios during DJF…”.

Line 321. “…Ziska2013-Mixed that include…”

Line 323. “…the maritime continent, which increases tropical…”

Line 324. “…and even more so in the MODERATE run where…”

Line 327. “…that can lead to entrainment…”

Line 329. “…occur frequently in this region in both seasons…”

Line 352. “…and 17 pmol L^{-1}...”

Line 405. “…concentrations to be between…”

Line 410. “…in the form of anthropogenic…”

Line 413. “…in this region and might explain some…”

Line 423. “…emissions with only slightly less bromoform (0.15–0.16 ppt) being transported into the UTLS…”

Line 436. “Desalination is mostly done in the Arabian Peninsula…”

Line 443. “…areas (Maas et al., 2019), respectively.”