

Interactive comment on “Importance of seasonally resolved oceanic emissions for bromoform delivery from the tropical Indian Ocean and west Pacific to the stratosphere” by Alina Fiehn et al.

Anonymous Referee #2

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Review of “Importance of seasonally resolved oceanic emissions for bromoform delivery from the tropical Indian Ocean and west Pacific to the stratosphere”, Fiehn et al.

General comments

This is an interesting paper that follows on from a series of similar studies by the same group investigating the sources of oceanic VSLs, their potential transport to the stratosphere and subsequent impact on ozone. The methods are mainly sound and have been reported previously so there are no major reasons that this paper cannot be published in ACP. Having said that there are a number of changes that I would like to see

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before I can fully recommend the paper for publication.

The paper is reasonably well written but there are many instances where the clarity could be improved. Parts of the document are quite difficult to follow and could do with some revision. I have highlighted some of these in the specific comments below.

The section on comparison with available measurement data (Section 3.2) is a little weak for several reasons. Firstly, for the comparison with ship measurements, why did the authors choose model output at 1 km altitude when the ship is sampling much closer to the ocean surface? As is shown in, for example, Sala et al (ACP, 14, 2014), there can be a large gradient in VMRs between the surface and 1 km, which could easily account for the differences shown in Table 3 and Fig S3. Is there a reason why data from the 2011 SHIVA-Sonne cruise (South China Sea) was not included in the comparison? Similarly, for the aircraft comparison, there are a number of other recent campaigns in the region covered by the model which would have helped to further validate the flux and model/transport calculations. Examples include SHIVA (Sala et al. 2014), CAST (Andrews et al., AMT, 2016) and ATTREX (Navarro et al., PNAS, 112, 2015). When deriving new emission scenarios like this it is worthwhile testing the output against as much observational data as possible.

Many previous studies have discussed stratospheric entrainment/source regions in the tropics and I am not sure you have done sufficient justice to this previous work. Comparison of your findings with some of these other studies should be considered.

I struggle a little with the overall conclusion of this study. The two emission scenarios seem to produce quite similar results when looking at the region as a whole but are strikingly different when it comes to the actual fluxes from the ocean and the location of these fluxes (e.g. Figure 2). Can the authors begin to address which approach is more realistic/promising and perhaps discuss what the key areas that need further research are. How do we begin to reconcile the large differences between inventories? Do we simply need more observations?

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Specific comments

L27-28 (also L93, L145, L154): be careful with the naming of the monsoon region. By Asian monsoon I presume you are referring to the Indian summer monsoon, rather than say the East Asian winter monsoon? Be consistent.

L35: I'm not convinced you can say that they "agree well" (there are significant differences in both the surface and upper troposphere comparisons). How about "agree reasonably well"?

L46: add "by", i.e. "vary by up to 50% ..."

L53: "they are of oceanic origin ...". Be specific – brominated VSLs are mainly oceanic but chlorinated VSLs are mainly anthropogenic.

L56: "Dorf et al and updates" – which updates are you referring to?

L56: "Uncertainties result from ..." I would argue that the uncertainty is also due to a lack of measurements of VSLs (both source and product gases) in the TTL and above.

L68-69: replace "As bottom-up approach ..." with "In the bottom-up approach ..."

L69-70: what is meant by "different spatial resolutions"? Do you mean ocean and atmospheric measurements in different locations?

L102: add "the" i.e. "Based on the first ..."

L102: "enhanced surface concentrations" – do you mean in the seawater or the atmosphere?

L108-109: should you add a date to the manuscript under review? Does the paper submitted to JGR differ significantly from this one (and Fiehn et al. 2017)?

L111: add "many" i.e. "... the topic of many global ..."

L119: change to "... only a few studies have considered ..."

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L122-129: Is it worth discussing what factors might affect the seasonality in bromoform sources here? What is the role of macroalgae relative to phytoplankton? The largest atmospheric concentrations are almost always near to exposed populations of seaweed. Do the emissions scenarios include this phenomenon? Annual changes in the tropics are presumably much less than at mid-latitudes and in polar regions?

L164-170: this section is a bit confusing. Are the new in-situ measurements (L166) from the OASIS cruise (L169)? L167: "These were used ..." What were used (the new inventories?) and where (in Fiehn 2017 or do you mean in this work?). This whole paragraph should be written more clearly.

L185: add "discussion" i.e. "in the following discussion"?

L187: move "in 2011" to the end of the sentence.

Figure 1 and Figure 3: it is difficult to distinguish between the different dashed lines. Can you try different line symbols?

Figure 1: Why are the atmospheric VMRs used in the 2 inventories so different? What would be the effect if both used the same atmospheric concentration? What impact does halving (or doubling) the atmospheric level have on the flux calculation?

L256-257: "We only calculate bromoform source gas injection to the stratosphere". Do you mean that you do not consider product gases at all? Perhaps you should state this for clarity?

L268: what drives the high emissions along the NH coastlines? Macroalgae? I guess you imply this later on (L 280-281) but why not state it here first?

L272: what is meant by "elevated atmospheric mixing ratios"? Where would the elevated levels come from?

L278: add "significant", i.e. "but show two significant differences. ..."

L279-283: If the Stemmler approach does not consider macroalgae and the effect of

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coastal processes then surely it will always underestimate bromoform emissions? How important are macroalgae relative to phytoplankton, particularly in these regions?

L287-289: It is not clear from Figure 2 that emissions are necessarily higher in winter and summer than they are in spring and fall. Can a more robust or statistical case be made (total flux from the region in each of the 4 seasons for example)?

Figure 2: What is the cause of the high winter emissions (in the Ziska inventory) from the Chinese and Vietnamese (and Philippines?) coastlines? This appears to be a strong source region that you do not really discuss in the text. Given the prevailing NE winds at that time of year this could be an important source of bromoform to the tropics (see, for example, Ashfold et al., ACP, 15, 2015 or Oram et al., ACP, 17, 2017)

Line 317: I agree that the coastal emissions are similar in magnitude but they are vastly different in location.

Table 1: If the numbers given are annual averages, what do the ranges shown represent?

Table 1: Are these numbers just for the Indian Ocean (i.e. not the full geographical area shown in Fig 2)? Please define what is meant by the Indian Ocean. Also I wonder if you should avoid using the term IO as it could be mistaken for iodine oxide!

L352: add “flux”, i.e. “To evaluate our flux and transport calculations”?

L352-353 replace “available” with “selected”, i.e. “from selected ship and aircraft campaigns”.

L356: should be “Table 3” not “Table 4”

L368: Begin sentence with “It is likely that oceanic sources. . . .”? Although please refer also to my general comments on Section 3.2 above.

L382-384: another, and possibly more likely (?), explanation would be the underestimation of the role of convection in this region. How well does FLEXPART deal with

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convection?

L405 —: In this section I think you should describe Figs 4a and 4b before discussing Fig 4c. As written, it is a little confusing.

L407-409: I am slightly confused by the term “transport efficiency” and how this was derived. In lines 258-260 it was defined slightly differently than it is here. How is the spatial distribution of transport efficiency independent of the emission scenario used when the mass emitted is different for the 2 scenarios? As I understand it, Fig 4c is a general picture which shows from which regions idealised particles will cross the CPT and has nothing to do with the bromoform emission inventories at all? If I am right, the term “bromoform delivery” in the Figure caption is misleading. A little clarification here would be appreciated.

Figure 4c. I am intrigued as to how the particles in the north east corner of the map get into the stratosphere during the summer months (JJA) when the prevailing winds in the region are from the southwest. Do they enter through the Indian monsoon or by some other mechanism?

L450: “Asian coastal areas” is a bit general. Which bit of Asia?

L454: I think you need to define again what you mean by the “stratospheric entrainment region”. Please explain clearly what is depicted in Figure 5 and how it differs from Figure 4. Does Figure 5 show the geographical location at the CPT where particles pass through to the stratosphere? If so, it seems odd that the southern tip of India is so important when I thought the main convection occurs further to the north?

L470 (Section 3.4): this section would benefit from a better description of the difference between transport efficiency and entrainment (as discussed above).

L491-493: the temporal shift is not particularly obvious from Fig 6c.

L497-500: This sentence is not very clear. How do the “differing annual cycles of bromoform entrainment to the stratosphere” influence the “regional pattern of entrainment

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to the stratosphere”?

L552 and L553: exactly 50% higher or approximately 50% higher?

L556: What altitude range does the anticyclone typically cover?

L615-617: “seasonality is only affected by wind speed and ss pressure”. Is that because the atmospheric and ocean concentrations are assumed to be constant throughout the year?

L617-619: “The Indian Ocean has a pronounced seasonality in ocean currents and upwelling regions (Schott et al., 2009) affecting the biological productivity, surface bromoform concentrations, and emissions”. Why include this sentence here? Do you mean to say that these are not included in the Ziska calculations? If so, please say so for clarity.

L645-649: This last sentence is not clear. What contributes “approximately half of the total stratospheric VSLS-Br”? Source gases in general? Where does the other 50% come from – product gases?

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