

Fuhlbrugge et al. have done a much better job of getting their main points across in this revised version of their manuscript, “Meteorological constraints on oceanic halocarbons above the Peruvian Upwelling”. The paper is clearer and, as I mentioned in my earlier review, the data look to be good and the overall approach sound. The paper now addresses uncertainties, the authors have removed much of the unnecessary text and clarified portions of it, and they have looked at upstream sources. That being said, I still have some concerns about the manuscript that warrant additional revision. This paper has potential to be a significant contribution to our understanding of how upwelling areas contribute to these gases in the atmosphere, but it needs to be written in a way that captures the essence of their findings.

#### General Comments:

I’m having a hard time coming to terms with the authors’ suggestion that the larger mixing ratios of these gases over upwelling areas are due to advection from somewhere else. Although they’ve shown some numbers using mean values and based on certain assumptions, they need to do more to convince me, and probably others, of whatever explanation they come up with in the end. Is the Peruvian upwelling all that different that it requires advection from somewhere else to elevate the mixing ratios? If so, how would one account for this in open-ocean upwelling areas? How much does the upwelling contribute to the excess, say, beyond what is found over the open ocean? The simple selection of advection for this area is not enough and I think the authors have enough information in their data and in the published literature to go further with this and draw out some generalities about air-sea flux and transport of short-lived halocarbons in the atmosphere.

Although improved in many places, the text still seems to meander or jump around, especially in the Discussion, but also elsewhere. In my view, some text is still unnecessary. Along those lines, while I fully appreciate the difficulties in dealing with the nuances of a second language, and the authors are overall doing a very good job with it (far better than I would do in German or Norwegian), I still find a number of statements throughout the paper that are awkward and require additional effort of the reader to understand just exactly what is being said. Since one of the co-authors is native to the English language, I would strongly recommend that he carefully go through the final version simply to ensure clarity and flow of the text before the manuscript is resubmitted.

The authors need to do a little more work on demonstrating their approach clearly, as their assumptions are key to the validity of their conclusions. Some of the text remains disjointed and needs tuning (see points below). Their treatment of statistics and uncertainties is improved, but they’ve not addressed representativeness of their choices and that is a bigger part of the overall uncertainty analysis. There are several good points to be made as a result of this paper – things that will improve our understanding of the contribution of upwelling zones overall to short-lived halogens in the atmosphere. This paper can come across strongly if it is organized about making those few points and can justify them.

The repeated comparisons to the Mauritian upwelling during the parts of the paper are distracting and they are not handled well in the Discussion either. It would be best for the authors to convey their findings for this study overall in a way that is clear to the reader, and after that have a separate section to compare to characteristics the Mauritian upwelling and other zones (noted below), but only to underscore their main points.

Finally, the paper has too many subheadings. That's an easy fix.

Specific Comments:

1. Abstract. In my mind, an abstract needs to say no more than “what we did, what we found, and what it means”. Any more should be reserved for the introduction or elsewhere. In this light, the authors could improve their abstract by deleting the first two sentences. The third sentence states clearly, succinctly, and compellingly what they did. It also identifies why this might be important (in this case, unique) and is a very good lead sentence. I offer a few minor suggestions below to help strengthen the abstract overall.
  - a. Lines 19-20. Awkward. Please rewrite, e.g., “oceanic emissions of dibromomethane and bromoform were relatively low, while those for methyl iodide were high . . .” or something to that effect. Also, “relatively” refers to what? Other parts of the ocean? Methyl iodide emissions? Etc.
  - b. Line 21 delete “both”.
  - c. Lines 24-27. Awkward sentence? Perhaps simply state that the specific properties correlated well rather than “correlations were found”?
  - d. Lines 26-27. Insert “below the trade inversion” after “abundances”.
  - e. Is the last sentence of the abstract true? Does it really reflect the authors' findings or is it conjecture? They make a case for emissions in their study area not being all that significant in delivery of the brominated compounds to the MABL, which seems counter to what this sentence says.
2. Introduction
  - a. Lines 34-37. This sentence seems to conflate tropospheric and stratospheric processes, e.g., the phrase “ozone depletion” normally is used to address a distinct phenomenon in the stratosphere. Try something like, “Oceanic fluxes of short-lived halocarbons contribute to reactive halogens in the atmosphere, where they are subsequently involved in ozone chemistry, aerosol formation, and other chemical cycles that influence the fate of pollutants and climate” or something to that effect.
  - b. Lines 43-46. Shouldn't this be a more general statement than one confined to the Mauritanian upwelling area? Isn't it generally understood that the composition of anything in the MABL is largely a function of oceanic sources and meteorology?
  - c. Line 46. Delete “Especially” at the beginning of the sentence. Since this sentence clearly is about Mauritanian upwelling, maybe there is probably a good way to make it a logical follow-up from a general statement in the (reformulated) previous sentence.
  - d. Line 49. Insert “high” before “atmospheric”; delete “also”.
  - e. Line 55. Make “waters” singular.
  - f. Lines 56-59. The way this sentence is written makes one think that the referenced studies were conducted in the Peruvian upwelling. Perhaps rewriting the sentence would help, e.g., “Given that others have found elevated levels of short-lived halocarbons over upwelling regions, it is likely (or ‘we would expect’) that the Peruvian upwelling zone similarly is a source of these gases” . . . or something like that.

- g. Lines 62-63. Delete comma after “Although”; insert comma after “. . . Liu et al.2013)”.
  - h. Replace “studies” with “have”.
  - i. Line 69. Insert “calculated” before “emissions”
  - j. Lines 70-71. Delete sentence.
  - k. Lines 76-80. Delete. Not necessary.
3. Data and Methods
- a. Lines 87-88. Delete “to collect . . . data”. It’s not necessary.
  - b. Somewhere in this short paragraph, the authors should refer to a figure of the cruise track. The best they have in this version is Figure 2a, which should suffice, but then Fig 2 would have to become Fig 1, should they choose that route.
  - c. Line 94. Insert “above sea level” or “above water level” after “height”.
  - d. Line 96. Delete “UTC time”; it’s internally redundant and also redundant with the parenthetic reference to UTC in the same sentence.
  - e. Lines 99-100. Is this sentence relevant to the paper? If not, I would delete it.
  - f. Line 107-108. Delete phrase beginning with “the convective . . . temperature”. The explanations are clear in the following text.
  - g. Line 114. Replace “the determination” with “determining the height (thickness?)”.
  - h. Line 116. Delete “during the ascent”; replace “whose” with “for which an”.
  - i. Line 118. Replace “this stable layer” with “its”; replace “depth” with “thickness”(?). BTW, since “stable layer” is italicized in this discussion, shouldn’t “convective layer”, e.g., in line 109, also be italicized?
  - j. Line 122. Since the section on relative humidity (2.2.1) is mainly about MABL height, I recommend deleting it as a separate sub-heading. Same thing for 2.2.2 on Line 135. Then combine the text logically.
  - k. Line 124-127. Despite the authors’ protest, I have a hard time thinking this statement is necessary, considering the likely readership. It’s their call; just noting my disagreement.
  - l. Line 127. Delete “therefore”.
  - m. Line 131. Delete “An”
  - n. Line 132. Delete “therefore”; also does elevated humidity always require a low MABL?
  - o. Line 137. I think the authors mean “not” permitted here?
  - p. Lines 135-142. What I gather from this section (which, as noted, does not need to be a separate section) is that either the same boundary layer height was used for all sampling areas that were in the coastal upwelling zone where sondes could not be launched, or (more likely?) that the equation was applied to the observed relative humidity and windspeed for each sampling site in the upwelling zone. Please rewrite to clarify.
4. Atmospheric VSLs measurements
- a. Line 144. Replace “were” with “was”; (“total” is singular). Insert “at” before “3”; replace “3” with “three” (typically done for single digit numbers); insert “intervals” after “hourly”; insert “above sea level” (“above water level”?) after “height”.
5. Oceanic concentrations and sea-air flux

- a. Line 154. Replace “3-hourly” with “every three hours”.
  - b. Line 158-159. I presume “precision” is 1 sd? Of duplicate samples? Subsamples? Injections of calibration gas?
  - c. Line 161. No need for subsection title.
6. Trajectory calculations
- a. Line 188. Replace “launched” with “initiated”, here and elsewhere, when referring to trajectories.
7. Oceanic contribution to MABL VSLs abundances –First, I want to thank the authors for making their approach much clearer. Figure 1, associated with this section, is very instructive and helps make clear the underlying assumptions and the acronyms used.
- a. I do have one problem with the schematic. Typically, mass balance is written as “rate change in mass” = “rate in” minus “rate out”. In that sense, if all terms are on one side of the equation, sources would be positive and sinks negative. The problem I have here is that, though advection is a source, it is being treated as positive on the wrong side of the equation. Were it on the same side as the other terms, it would be negative and the concept would not make sense. So I recommend that the authors show (and explain) this either with all terms on one side of the equation and zero (i.e., “rate change in mass” for steady state) on the other side, or sources on one side and sinks on the other. The authors have correctly done this in their calculations, since the numbers for ADR in Table 2 add up correctly, but it is not really correct for AD in this figure mainly because of the sign. At best, it’s a little confusing as presented. (The colors help, however.)
  - b. The primary conclusion of this paper depends heavily on computation of the convection term. (And there is some discussion of it in sections 3.4 and 3.5.) It provides a large number that has a large uncertainty. I note the fluxes shown for this term shown in Table 2 and hope they are representative. And I hope the uncertainties also are representative and include the various terms that go into their computation, as they are probably not normally distributed. It might be instructive to see the distribution of this term for some reassurance, or for the authors to examine how departures from the mean values shown here or variability of convection throughout the region might affect their main conclusion and share that with the reader. If this seems picky, I only focus on it because the ultimate sources of these gases being advected into this region are left to some degree of speculation that’s a little hard to swallow. As mentioned in the general comments, it’s hard for me to think why mixing ratios are elevated over areas of upwelling worldwide, if upwelling isn’t contributing significantly or even predominantly to what is in the atmosphere. And I have a hard time thinking that Peru is all that special of a case as to warrant other sources. Although Peru is close to being coastal, there are other upwelling areas not close to the coast that still support high mixing ratios of these gases. So what’s going on? The answer to that could be very useful for future investigations and I think they have it in their data and in analyses they have not shown.
  - c. Line 195-196. Delete the second and third use of “to”; delete “the time scales of” before “air” and “the” before “chemical”; add “rates” after loss on line 196.
  - d. Line 206. Delete “process” after “this”.

- e. Line 211. Delete “acts as a loss process as well and”.
  - f. Line 212. Reference to “steady state” should be at the beginning of this discussion. It is a necessary assumption underlying all of this section.
  - g. Lines 219-222. See “a” above. Be sure this is right.
8. Observations
- a. Line 231. Change “backward trajectories” here and everywhere else to “back-trajectories”.
  - b. Line 231-232. Awkward sentence. Try “. . . reveal a mix of open ocean and coastal air-masses”.
  - c. Line 238. Change “is” to “was”; delete “for”
  - d. Line 242-243. Replace “The decreasing . . . upwelling” with “and indicating”.
  - e. Line 248. Replace “for example . . . Figure 3c” with (e.g., Dec 9-11, 19; Figure 3c).
9. VLSL observations and oceanic emissions – remove reference throughout to Mauritian upwelling and focus on results from this study.
- a. Line 254 (and elsewhere). Spell out single digit numbers.
  - b. Line 258. It’s a little unusual to rely to any degree on a paper that has only been submitted.
  - c. Line 259. Change “are” to “were”.
  - d. Line 260. “Low” compared to what? Focus on the temporal pattern. Stick with these results.
  - e. Line 268. Replace “and” with “to”.
  - f. Line 269. Delete “fresh”.
  - g. Line 270. Replace “are” with “have been”.
  - h. Lines 271-272. The authors use different lifetimes earlier in the text. Decide which to use.
  - i. Line 271-274. Refer to Carpenter et al. parenthetically at the end of the sentence rather than the middle; insert “recently” before “enriched”; delete “with fresh”.
  - j. Line 274-277. Delete “A general decrease of”; replace “the” before “concentration” with “This”; replace “is found” with “generally decreased”; delete “during the cruise”. Last phrase of the sentence is not clear. Is the reader to presume south? Also, what do they mean by “remote” – do they mean “open ocean”?
  - k. Line 280. Replace “synchronous” with “approximately simultaneous” or “approximately synchronous”.
  - l. Line 283. Replace “are” with “were”; what is meant by “low”?
  - m. Line 286. Replace “are” with “were”.
10. Lower atmosphere conditions
- a. Line 291-293. Replace “The relative humidity shows a” with “A”; insert “positive” after “strong”; insert “of relative humidity” after gradient. Also where does the 75% refer to? Delete “with height due to suppressed mixing.”
  - b. Line 304-305. Delete “are good indicators . . . parameters”.
  - c. Line 306-307. Delete sentence. This has been said already.
  - d. Line 316-318. Last phrase of sentence beginning with “before” is awkward. Revise.

- e. Line 308-313. Beginning with “The regressed . . . “. The important point here is what? Hasn’t it been made already? Is the range of the MABL heights something that could help explain their findings? Is it really correct to use the coefficients derived from open ocean radiosondes on something that yields a 10 m height?
  - f. Line 316-318. Last phrase in this sentence is awkward. This is a concluding paragraph, in that the authors state it as their interpretation. It needs to be clear.
11. Contribution of oceanic emissions and meteorological constraints (sections 3.4 and 3.5). These sections still seem a little long and rambling. Perhaps they could be shortened. They certainly meander. Maybe a little reorganization focusing on the impacts of the uncertainties or variability would help? Also, move the comparisons to Mauritian upwelling to a separate section, as noted in my general comments.
- a. Line 328-329. “a mean residence time for FLEXPART trajectories of 7 hours in the observed MABL during the cruise”. What exactly does that mean? At a velocity of 10m/s, which works out to 36 km/hour, it would seem these trajectories go back (or are stable within?) ~250 km or ~2-3 degrees latitude? I’m not sure why 7 hours was chosen and I’m sure others might be confused as well.
  - b. Line 331. Change “origins” to “originates”.
  - c. Line 331-332. 3% + 99% = 102% . . . . please fix or else explain.
  - d. Line 335. The authors might consider putting comparisons with South China and Sulu Seas in with a Discussion section on Mauritanian upwelling, i.e., how results from this region compare with other parts of the ocean.
  - e. Line 337-339. Sentence fragment.
  - f. Line 342. “coastal” sources . . . I presume this means outside of the study area?
  - g. Line 364-369. Has this been said already? I think much of it has.
  - h. Line 375. “co-correlated with each other” is redundant.
  - i. Lines 380 ff. M91? DRIVE? Readers might need a little help here.
12. Discussion and Summary – Sections 4 and 5
- a. I think this section has much of what I’m asking, but it rambles a lot and inserts bits of speculation throughout. It’s hard to walk away with a conclusion, or even any clear points in my head. What will help most is for the authors first to interpret the results that have been described, including the part addressing uncertainties in their data and assumptions, then go ahead and start making comparisons, but only where they are important and support the main conclusions. Support statements quantitatively where that can be done. The authors need to convince themselves about what points they want the reader to take away and then organize the discussion around them. It would make for a more enjoyable and more informative read.
  - b. The summary can be tightened up considerably following a reorganization and rewrite of the Discussion. If the discussion is well done, then a summary might not even be necessary.
13. Figures
- a. Figure 1 – (see comment above). Also “summery” in the legend should be “summary”.
  - b. Figure 2 – this, too, is a very helpful figure
  - c. Figure 3 – nicely done.

- d. Figure 4 – what is a “specific altitude” (line 570)?
  - e. Figure 5 – Line 587. Insert “near surface” in front of “atmospheric”
14. Tables
- a. Table 1 – Line 558. Uncertainty: which is it, s.d. or range? They are different.
  - b. Table 2 – aside from my previous concerns noted above about the representativeness of the values shown here, this table’s OK.