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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Air-sea fluxes of biogenic bromine from the tropical and North Atlantic Ocean" *by* L. J. Carpenter et al.

Anonymous Referee #2

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General Comments

The manuscript by Carpenter et al. presents air and seawater measurements and sea-to-air fluxes of two halocarbons (CH2Br2 and CHBr3) from the Atlantic Ocean. The authors compare the influences of coastal and open ocean emissions on the distributions of these gases and assess the importance of these different regimes to the global budgets for these two compounds.

Overall, the manuscript presents new measurements and fluxes for these gases in two different regions of the eastern Atlantic. The authors address the global budgets for these gases, but more importantly show from their measurements that the eastern Atlantic was supersaturated throughout the sampling regions and phytoplankton abun-

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dance may influence levels observed (ultimately in the atmosphere) outside of coastal regions. Nonetheless, this work illustrates that considerable efforts are necessary to better constrain and quantify the global budgets of CHBr3 and CH2Br2.

The scientific methods and assumptions are valid and clearly outlined. However, it is unclear if it the style of the journal to only provide the manufacturer name when identifying a specific piece of equipment or parts in the experimental section? For instance, the authors refer to using a "clean metal bellows pump" and only cite Aerospace Ltd. - it would be helpful if model s and company locations were also included. For example, referencing a specific metal bellows pump, I would include the following information: (MB-302mod, Senior Flexionics, Sharon, MA) as opposed to just listing "Aerospace Limited". While the authors reference other work regarding experimental details, it would be useful to the reader if the pertinent details regarding the flux calculations were provided in the manuscript. As the paper currently reads, there is nothing that outright states that the simultaneous measurements of the sea surface water concentrations, atmospheric mixing ratios, water temperatures and instantaneous wind speeds were used to estimate their fluxes...assuming this is what they did.

The results are sufficient to support the interpretations and conclusions drawn in the manuscript. However, it would be much more useful to the greater community if a larger array of data were included in sections 3.3 and 3.5, especially because the title of section 3.3 includes the following "...fluxes and comparison with literature" - there are an array of other recent measurements and fluxes for these gases reported in the literature which go beyond the Butler et al. (2007a) paper. I feel this would better reflect the uncertainties and ranges in the different coastal regions as well as begin to address the influence of seasonality on emissions. Other more recent work should be used and cited throughout the manuscript, especially sections 3.3 and 3.5.

Is the overall presentation well structured and clear? Yes, with the exception of the figures - please see the specific comments section below.

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Is the language fluent and precise? Yes, with a few minor issues addressed in the specific comments below.

Are mathematical formulae, symbols, abbreviations, and units correctly defined and used? In the figures the units of the fluxes are presented correctly, but not in the text - should be nmol m-2 d-1. Also, as stated previously, it would be instructive to the reader if the important details regarding the flux calculations were included.

Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated? The quality of the figure presentation needs to be greatly improved. Font sizes are too small, scales are not comparable; overall it is extremely difficult to read/extract the information out of the figures in their current form.

Specific Comments

Abstract (P 18410) L6 - this sentence should reflect that the order is specific to this work L8 - need a space between "chlorophyll" and "a" L9 - "coastally-influenced" needs to be defined, quantitatively or semi-quantitatively in the abstract such that the budgets be put in to better context. Also, only CHBr3 s are presented in the abstract - CH2Br2?

P18411, L5 - delete "especially CHBr3"

P18411, L13 - replace "fuller", inappropriate word choice

P18413, L16-18 - revise sentence, not sure what you are trying to convey, also need a "(" before Peterson citation

P18413, L19 - replace "taken" with something more concise such as "collected and analyzed"

P18414, L9-12 - the resulting r2 vale of 0.28 implies that there is no correlation with Chl-a; this should be removed from the text. The best that can be said is that there may a loose association, but there is nothing to quantitatively substantiate this statement.

P18415, L9 - same issue regarding the r2 value w/ Chl-a - remove from text.

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P18415, L15-18 - revise the following: "were similar to measurements in waters of between 20-45m depth" to something like "were similar to surface water measurements at depths from 20-45m at Mace Head..."

Also need a space between "at" and "depths" after 68 pmol dm-3

General comment for section 3.3; it would be useful to include other data here and possibly include a table summarizing results from other studies (Quack and Wallace, Quack, Chuck, Zhou et al., etc.,) in addition to Butler et al.

P18417, L1 - delete "that" preceding "in coastal regions" P18417, L2 - delete "the" preceding "light of their..."

P18417, L4-7 - regarding the following statement "Atmospheric bromocarbon ratios have been used to deduce global estimates of their fluxes (Carpenter et al., 2003; Yokouchi et al., 2005) with an implicit assumption that the oceanic emission ratio is near constant."

A noteworthy detail illustrated in Zhou et al. [2008] is that when CHBr3 and CH2Br2 mixing ratios at Appledore Island, ME (Gulf of Maine, western Atlantic) were above about 20 pptv and 5 pptv, the ratio of these two gases deviated from a linear relationship through their data set. While these results aren't specifically unique, they do indicate that different factors may effect direct emissions from the surface seawater and that the oceanic emission ratio is not constant. Factors such as this should be incorporated in to the discussion that "coastal measurements should be treated with caution when inferring global average emission ration.", as this assumption is only a first order approximation, at best, and clearly, measurements show that the oceanic emission ratio is not constant. It would be quite instructive if the others addressed how different emission ratios would impact their global budget estimates. Moreover, the difference in the slopes presented Zhou et al. for these different emission "regimes" may provide additional insight regarding sea-to-air transfer, air mass processing and transport.

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P18418, the end of section 3.4 ends rather abruptly - it would be beneficial if the authors concisely summarized the key points of this section.

P18418, section 3.5 - other data sets should be included in the global and regional flux estimates such that a representative range from a more comprehensive set of measurements is presented. Also, the authors comment on the use of bathymetry data to determine the geographical area of coastal regions and that depth profiles are useful, other coastal region sources are not addressed. For completeness, other sources should be addressed in the discussion.

P18418, L24 and 26 - fix units on fluxes (should be m-2)

General comment on Figures - all figures and fonts should be made larger; in their current form, it is difficult to read the axis units, labels, scales, etc.

Figures 1 and 5 - For ease of comparison for the x and y scales should be the same for the halocarbon data and the Chl-a image (ideally, all three figures would be uniformly sized).

Figures 2 and 3 - it is difficult to see the features of the middle plot (Chl-a, temp anomaly and log depth) - either use more clear symbols/lines for each, different colors or scale differently. Also, in Fig 2, there is one negative flux point and several near zero points - these aren't addressed in the text.

Figure 4 - it is not obvious that Fig 4 is the best way to illustrate that higher concentrations are observed in the SACW. Qualitatively, what does the fact that the slopes of the lines denoting SACW and NACW are similar? While they appear to have been included to guide the readers eye, are the slopes different? If so, is this difference significant? Also, what is the frequency distribution of the CHBr3 and CH2Br2 in each of these regions? From the first two panels (CHBr3 and CH2Br2), it appears that the range of distributions are comparable - further details would be useful. However, the delineation of Chl-a between regions is quite apparent. The fact that there is only one

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sentence in the text referring to this figure makes it questionable if it is beneficial for inclusion.

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