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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Global sea-to-air flux climatology for bromoform, dibromomethane and methyl iodide" *by* F. Ziska et al.

Anonymous Referee #2

Received and published: 24 April 2013

This paper attempts to derive global flux estimates of three, short-lived halogenated gases from perhaps the most extensive atmospheric and ocean observational datasets available. Though the extensive data are still sparse when viewed on a global scale, they are extrapolated in space with two different techniques to derive global values. There are substantial uncertainties and shortcomings associated with the analysis, such as the sparseness of data, inconsistent standardization, potential seasonal biases, and a poor process-based understanding that make extrapolation (or interpolation) difficult, but this is true for all studies of this ilk. The authors are clearly aware of these issues, and attempt to do the best job possible to provide an observation-based estimate of global fluxes and their distribution. As a result, I think this is a useful analysis, as it provides a unique, observation-based estimate of these fluxes derived from much of the globally-available data that will be important to consider in future global

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Interactive Discussion

Discussion Paper

modeling analyses striving to better understand the impact these emissions have on the global atmospheric environment. I have only a few comments to improve clarity and accuracy.

There are instances where over generalizations of interpretations are provided, specifically when describing similarities between different chemicals. For example, some have suggested that CH2Br2 and CHBr3 share common pathways for emission, etc., and in a number of instances (e.g., lines 374, 434, 440) the authors comment on the similarity in concentration or flux distributions for these chemicals. But in many instances I have to disagree with the authors and suggest that the distributions for these two gases often appear quite different, despite their assertions otherwise. I'd recommend that the authors avoid over-general comments related to similarities in distributions, and instead focus their comments on specific regions.

Section 5.2, I think conclusions about seasonally-varying fluxes are highly suspect, given that only annual mean air and water concentrations are being considered here. These seasonal variations are only driven by seasonal changes in the variables driving exchange, if I'm reading the manuscript correctly. It would seem that seasonal changes in water concentrations are highly likely, and, therefore, I presume that such variations could significantly alter our understanding of how the fluxes actually vary with season. I recommend removing this discussion or very clearly emphasizing in this section that these conclusions regarding seasonal variations in flux do not consider one critically important part of the equation (seasonal concentration variations).

Section 5.3 is interesting, but needs some comment on the amount of data going into the climatology in this region; is the agreement the result of their being very little data other than that from the Quack et al., 2012 cruise going into the climatology here?

Regarding timescales and potential influences of covariations in exchange and concentrations on flux estimates, the information on line 603-606 needs to come in the methods section of the manuscript, not in results and discussions. Regarding this (and

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the point made above regarding section 5.2), are there no longer-term measurements that help one assess seasonality for these gases in water or marine atmosphere that could potentially be extrapolated to broader regions as a sensitivity test?

Differences between the two regression techniques are described, and the ordinary least squares is indicated correctly as being more influenced by outliers. One must remember that the true emission magnitude could be driven by the outliers, to some extent, and even the OLR technique would underestimate the influence of those outliers (e.g., line 685). Hence I'm not convinced that it can accurately be described as an upper limit estimate (as you indicate on line 673)... some discussion of this is given in lines 590-592, but be specific about which study you mean by the work "there" (line 592).

Details: Not clear if fluxes in sampled grids are from observations or from regression results (which are used for non-sampled grids)...

Lines 688-690 conclusions here don't appear to be any that can be drawn from evidence provided in this paper... I'd recommend removing them or attributing them properly.

Units on RMSE in tables C1 and C2?

Seems to me that the first two paragraphs on p. 17 make the same points, perhaps one was meant to be deleted?

Quote fluxes in comparable units throughout the paper and in Tables and figures (moles or grams of halogen or compound...)

Standard of English needs improving.

Line 127, I'm not convinced that iodine affects stratospheric ozone. Lines 382-387, are these differences robustly determined by measurements in multiple seasons and/or from multiple groups so that we can be sure they are the result of one groups unique calibration standard scale or measurement bias, for example?

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Interactive Comment

Is there any potential explanation (other than the obvious but not all that informative one given) why the Arctic and Antarctic should give quite different CH_2Br_2 fluxes?

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 5601, 2013.