

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

**Anonymous Referee #1**

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The study of Ziska et al. provides an important and timely global bottom-up climatology of sea-to-air flux estimates for bromoform, dibromomethane and methyl iodide. This data set will greatly help to better constrain the emission of very short-lived halogenated compounds in global atmospheric models. The paper is generally well written and I recommend publication in *Atmos. Chem. Phys.* after consideration of the following comments.

### General comments

The constructed climatologies constitute a very important and useful data set. As discussed in the text, the available observations are still sparse and much of the analysis is related to filling the gaps by some kind of inter- or extrapolation. While the general approach followed here seems very reasonable and justified, I have some comments

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and suggestions regarding the interpolation procedure that should be considered:

1. The in situ observations are mapped on a global 1x1 degree grid through a regression with latitude and longitude to construct a background field as described in sections 3.4 and 3.5. If observations are available within a 1x1 degree grid cell, these data are instead taken from an objective mapping technique (section 3.3, if I understood this correctly, this point wasn't made fully clear). As a result of this different treatment, there are structures in the climatologies, that represent the availability of data, not real features (seen e.g. in Fig. 4). While I understand that this approach has been chosen to provide the best possible available information, I still find this problematic for the use in global models. My suggestion would be to make available different tiers of the data sets: E.g., Tier 1 could contain only the incomplete data from the objective mapping (similar to Fig. 1), Tier 2 could contain the results from the linear regression with latitude and longitude only, while Tier 3 could contain the merger of both, as for the current data set. Anyway, I would encourage the authors to make these data publically available as a supplement to this paper.

2. While it seems very reasonable to use different latitude regions for the oceans and the atmosphere, there is the risk that this introduces some inconsistencies for the sea-to-air concentration gradients. E.g., consider a hypothetical situation where concentrations in ocean and atmosphere are in equilibrium but each change with latitude. Calculating regressions over different latitude regions for ocean and atmosphere could then introduce artificial sea-to-air concentration gradients. I am slightly concerned that the pronounced concentration gradients for bromoform seen at about 60S (minimum) and 60N (maximum, Fig. 6) could be related to the particular choice of latitude ranges for the regressions. I suggest to double-check that these features are real and robust and not an artefact due to the regressions used.

3. Given the importance of emissions from coastal and shelf regions compared to the open ocean, I feel that more details should be given how coastal and shelf regions are defined and how sensitive the results are to the particular definition. Is the shelf just

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the region 2 degrees away from the land, or is it based on water depth or some other parameter? Also more information is needed if and how the coastal and shelf data are further divided into different latitude regions (Sec. 3.2). Most of the current discussion is limited to open ocean.

Specific comments

p.5603, l.6: "In input of ...": make clearer that this refers to previous studies, not the current study.

p.5603, l.18: "global concentration gradients": you mean sea-to-air concentration differences?

p.5604, l.20: I suggest to move this sentence towards the end of the paragraph

p.5606, l.19: remove parentheses around "bromoform, ..."

p.5610, l.1: "The data classification...has been linked...": what specifically does that mean?

p.5610, Sect. 3.3: More information is needed here or should be made clearer. There is a correlation length for the Gaussian weighting (3 degrees in open ocean, 1 degree for shelf and coast) and also a cut off length? Sorry, this was confusing to me – and may be relevant for my general point 1 above.

p.5611, l.13: remove the word "vertical"

p.5611, ll.15-20: please provide more information for the RF method.

p.5611, last paragraph: in addition to the RMS you may also consider the uncertainty for the trends. If the uncertainty is large, it may be better to use just a mean of median, instead of a trend. By the way: Have you considered a continuity criterion for the trends in adjacent regions, or could there be sharp breaks between adjacent regions?

p.5612, Sect. 3.6: to fully appreciate the functional dependence of the flux on wind

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speed, SST, pressure and salinity it would be helpful to have also the equations included how the equilibrium water concentration  $C_a$  is related to atmospheric mixing ratios (via pressure) and how the Schmidt numbers depend on temperature and salinity. This would then help to better understand the relationships shown in Fig. 3.

p.5612, eq. (2): This is not dimensionless, i.e. it depends on the units for  $U$  (I guess m/s). Provide units or (if possible) write in dimensionless form.

p.5616, l.27: "as reported in the literature": replace by relevant citation

p.5619, l.24: in what sense is this an "upper limit"?

p.5621, l.21 and 27: do you interpolation or extrapolation?

p.5624, l.11: "less than 1%": I don't understand. 1% with respect to what? And why?

p.5625, Sect.5.3: Are TransBrom data part of the climatology, or is this a totally independent data set? Other point: is this really "short time" variation, or also small-scale?

p.5626, l.6: missing "."

p.5633, l.27: If Quack et al. (2013) is still "in preparation" it should not be included in the refereneces list.

Figs. 6 and 7: The concentration gradients in Fig. 6 look surprisingly similar for RF and OLS. One can identify differences, but overall the differences are small, even for bromoform. With that I have difficulties to understand why the sea-to-air flux as shown in Fig.7 is almost a factor of 2 larger for OLS than for RF for bromoform.

Fig.7: Can't see any "gray shaded" area.

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