

Interactive comment on “Marine sources of bromoform in the global open ocean – global patterns and emissions” by I. Stemmler et al.

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The authors implement a bromoform module into a global ocean circulation model (MPIOM) coupled to a biogeochemical model (HAMMOC). They investigate the impact of planktonic production and lateral transport on bromoform surface concentration patterns and on gas-exchange with the atmosphere. They also evaluate the model against observations. Overall the model is capable to represent large-scale features of observed bromoform concentrations. The analysis of some sensitivity simulations suggest that a better match with observations might be achieved by reducing bromoform production in the model.

This is a very nice piece of work, with implications for both the ocean and atmospheric communities as well as for Earth System Modelling. As indicated by the authors, there

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is a great potential for coupling this ocean model to an atmospheric model to investigate the impact of bromoform processes in the ocean on the exchange with the atmosphere and subsequent atmospheric transport (although the ocean-to-atmosphere fluxes reported in this study look too low compared to previous estimates). I encourage the authors to bring this further in the future, try to represent coastal sources and even consider the ocean modelling of another important source of Br to the atmosphere (CH_2Br_2).

Results are generally discussed in an appropriate way and the authors give credit to previous work. However the use of English is not always appropriate and the manuscript is not carefully finished. There are indeed a large number of typos which make the text unnecessarily difficult to follow. In particular, the authors have done a poor job when referring to the figures (both those in the main text and in the supplementary material).

Since this work is a substantial contribution towards scientific progress in the field I recommend publication, but only once the comments below have been properly addressed. There are some relatively minor scientific issues, but a large number of technical corrections are needed before this manuscript can be published.

SPECIFIC COMMENTS

(1) It would be convenient to give further information about the Ziska's dataset used in this work. Two examples of how the text could be improved:

1.1. Towards the beginning of Section 2.2 (Model set-up) the authors write "All of these experiments use the climatological atmospheric concentrations of Ziska et al. (2013) as upper boundary conditions" but they do not give many explanations about that dataset. Then at the end of the first paragraph of Section 2.3 (Observations) they elaborate a bit more on this: "... Ziska et al. (2013). The gridded atmospheric mixing ratios from the robust fit method are used as boundary conditions for the model after conservative spatial interpolation onto the model grid". Since all these details are relevant for the

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model set-up, why not including them in Section 2.2? Or at least mention there that more details will be given in the following section. Also the authors could clarify why they use Ziska's data from the robust fit (RF) method instead of from ordinary least squares (OLS) regression. Also note that it is convenient to indicate what RF and OLS stand for in any of these two sections since such acronyms are used but not explained later in the text (e.g. Table 3).

1.2. Section 2 (Model setup), second paragraph in page 15701: "The additional experiment, Seas-at, differs from Ref only by the atmospheric boundary conditions for bromoform gas-exchange. Here, a monthly mean annual cycle was imposed onto the atmospheric boundary conditions, which was derived from the annual cycle of surface ocean bromoform concentrations simulated in Ref". Then the authors justify how they do this. Could you clarify whether the data from Ziska is available on a monthly basis? And if so why did you impose your own annual cycle? I will go back to the temporal variability of Ziska's dataset in another question below.

(2) At the beginning of Section 3.1 the authors write "The spatial distribution of bromoform in seawater reflects the balance between sources (production and uptake from the atmosphere) and losses via outgassing and degradation". Three of those processes (uptake, outgassing and production) are included later in Table 2 while degradation is missing there. Then they write on page 15705: "At the global scale it is dominated by gas exchange (~250 d, Table 2), the residence time with regard to degradation is much longer (~1100 d, Table 2)". How can they give a residence time related to degradation and refer to Table 2 when that process is not included there? And what do the numbers in brackets at the end of each column of that table mean?

(3) Page 15710, last sentence of section 3.2: "The best match with observations is achieved when either reducing the bulk bromoform production rate, or considering lower bromoform production by diatoms than by non-diatom species (Fig. 6)". Having a look at the figure I am not completely convinced that the Dia simulation (pale purple) matches the observations better than Ndia (pale red). It looks like there is

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some dependence on the geographical location, which is not surprising since diatoms prevail in high latitudes and non-diatoms in low latitudes. Can the authors do a more careful evaluation of this?

Also, since that last paragraph is a nice summary of the whole section 3.2 I would recommend including it in a new sub-section. Otherwise it looks like it is part of the "Southern Ocean and Arctic" subsection.

(4) page 15712, lines 4-9: "They include three top-down inventories (Warwick et al., 2006; Liang et al., 2010; Ordóñez et al., 2012), and the bottom-up (based on observations in air and water) inventory by Ziska et al. (2013) (OLS method). They are able to reproduce most of the seasonality of bromoform atmospheric mixing ratios with these temporally invariant emissions, presumably because it is driven by photolytic degradation in air (Hossaini et al., 2013)". I had a look at those papers. Emissions might be temporally invariant for two of them (Warwick et al., 2006; Liang et al., 2010), but I am not sure that is the case for the other two:

* Ordóñez et al. (2012). See section 4.2 (Implementation of VSL halogenated sources in CAM-Chem). A monthly climatology of Chl-a is used to produce emissions of bromo- and iodo-carbons. The formulation of the fluxes between 20 N and 20 S is: $E = 1.127e105 \times f \times r \times chl-a$. Since this is proportional to chl-a, which experiences some variations from month to month, there should be some temporal variability in the distribution of emissions over any latitudinal band.

* Ziska et al. (2013). They calculate global monthly sea-to-air flux averages of bromoform (see e.g. Fig. 8, but also other parts of the text), although I can also read that they calculate some annual climatology. The authors know that dataset much better than me so they can clarify that.

(5) Table 3 does not seem to be accurate. Please have a look at all references and then do your own calculation of the bromoform fluxes in the appropriate units (instead of getting the values from Ziska's paper which might contain some errors). I believe

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that some units in that table are Gmol Br yr⁻¹. You only indicate Gmol yr⁻¹, but it is not clear whether you mean Gmol (Br) yr⁻¹ or Gmol (CHBr₃) yr⁻¹. I did a few checks:

* Liang et al. (2010) reported 425 Gg(Br) yr⁻¹ → this is ~5.32 Gmol(Br) yr⁻¹. Very similar to the value you show but you need to indicate the right units.

* Ordóñez et al. (2012) reported 533 Gg (CHBr₃) yr⁻¹ → this is ~2.11 Gmol (CHBr₃ yr⁻¹) or ~6.33 Gmol (Br) yr⁻¹, somewhat lower than the 6.67 value you show

* After some sensitivity simulations, Warwick et al. (2006) reported the range 400–595 Gg (CHBr₃) yr⁻¹. I haven't tried to do the conversion, but you give a single value without explaining why.

* Your references to Ziska's fluxes seem to be right (1.5 with RF method and 2.49 with OLS method) as long as units are Gmol(Br) yr⁻¹ as indicated in that paper

* Then on line 4 of page 15711 you say that the global flux from this work is around "0.3 GmolCHBr₃ yr⁻¹ (Table 3)". Is that correct? If so you would have 0.9 Gmol (Br) yr⁻¹, which is the value you should show in the table if you are working with those units. I am aware that your flux might be lower than previous estimates partly because you don't intend to represent coastal emission. However if it is too small and you couple the ocean model to an atmospheric model in the future then the CHBr₃ loadings in the atmosphere will most probably be too low.

Please be careful with the units and revise the whole Table 3. Then check any flux values reported in the main text and any conclusions you make when you compare them.

(6) It would be good to include a clearer discussion in the last section (Conclusions) about the net negative fluxes of bromoform at high latitudes. This was also found by Ziska et al. (2013) and it is a very relevant result since other studies did not consider that possibility (e.g. Warwick et al., 2006; Liang et al., 2010; Ordóñez et al., 2012). The authors should discuss whether potential issues in the model (e.g. missing bromoform

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production from sea-ice, underrepresentation of coastal emissions) might have some impact on those negative fluxes as well as on their low global fluxes compared to other studies.

(7) Finally, I find it hard to read the colour bars and text in Figures 1, 2, 4 and in particular Figure 7. The authors might consider re-arranging the panels (by increasing the number of rows and reducing the number of columns; but if they do so they have to take care to refer to the figures correctly from the main text). Please at least make sure that the text in those figures is readable when you get the proof-readings!

TECHNICAL CORRECTIONS

* page 15694, lines 24-25: "... one of the most abundant bromine containing volatile halocarbon and is one considerable ...". Change to "... one of the most abundant bromine containing volatile halocarbons and is a considerable ...".

* page 15695, line 20: References (Moore 1996; Hughes, 2013). The first one should be Moore et al., 1996. The second paper is missing in the reference list.

* page 15697, line 15: Change "species-(or group)-specific" to "species- or group-specific". Or at least leave space before/after opening/closing brackets.

* Ideally all equations should be followed by an identifier, i.e. (1), (2), ..., but I don't know if that is required by this journal.

* page 15698. Commas in the three following sentences should be removed: "Hense and Quack (2009) show, that ..." "As it was shown for freshwater nitrifiers, that ..." "As both processes are strongly temperature dependent, and follow different ...

* page 15699. You wrote: "We modified the description of the transfer velocity to (Nightingale et al., 2000): ... (formula) ... to resolve the temperature dependence of the Schmidt number ...". Better write something like: "We modified the description of the transfer velocity given by Nightingale et al. (2000) to resolve the temperature dependence of the Schmidt number ScCHBr₃ (Quack and Wallace, 2003):" Then show

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all the formulae. Also indicate that "u" in the first formula is wind speed.

* page 15701. When describing the model simulations you wrote "preindustrial conditions ($p\text{CO}_2 = 278 \text{ ppm}$)" and "present-day conditions ($p\text{CO}_2 = 353 \text{ ppm}$)". $p\text{CO}_2$ should refer to a partial pressure of CO_2 in the atmosphere while you are giving values of atmospheric mixing ratios. Please re-write.

* page 15702, line 15: Change "Chl a" to "Chl-a"

* page 15703, lines 4-8: "A reduction of the bulk production ratio of bromoform relative to primary production (Half) leads to a reduction of bromoform concentrations almost everywhere, apart from regions with uptake of bromoform from the atmosphere (e.g. the Southern Ocean and the northern extratropics in the local summer seasons, Fig. 1c and f)". The plots are small and therefore I find it hard to distinguish things, but I think you need to change "local summer seasons" to "local winter seasons" in that sentence.

* page 15703, lines 12-16. The following sentence is poorly written and needs to be improved: "Similarly, differences between Ref and Seas-at are highest, where a strong seasonal cycle in production results in a strong seasonality of CHBr_3 surface concentrations and by construction of the seasonally varying atmospheric forcing field of Seas-at, e.g. in the extratropics (Figs. 1, 7b and f around 50 N)". If possible split it. And you might need to mention Figs. 1b, 1e, 7b, 7f.

* page 15704, line 3: "As mentioned above, bromoform distribution patterns for the main part follow ...". What do you mean by "for the main part"?

* page 15704, line 9: "This distribution of diatoms is in line ...". Remove "of diatoms" from this sentence. I believe you refer to both diatoms and non-diatoms.

* page 15704, lines 16-19: "As diatoms dominate in productive regions, the impact of reducing the bromoform production rate by diatoms on the global CHBr_3 inventory is similar to the impact of reducing the bulk production rate by the same factor (Table

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2)". I understand what the authors mean, but is this clearly reflected in Table 2 when comparing data for the columns Half, Dia and NDia? You may need to remove the reference to that table, rewrite this sentence or explain this a bit more.

* page 15704, line 19: focusing onto → focusing on

* page 15705, lines 18-20: "As expected, lateral transport from shelf regions is particularly relevant in the Arctic surface ocean (Figs. 4c, f and 5), because of its hydrographic features (Mediterranean sea) and low outgassing at cold temperatures". This sentence looks strange. Need to re-write it to completely separate the Arctic and the Mediterranean, where conditions can be very different. This process is clearly relevant for the Arctic while the impact on the Mediterranean seems to be important during boreal winter (Fig 4b). Also the authors might mention "(Figs. 4b, e and 5)" instead of "(Figs. 4c, f and 5)".

* page 15705, lines 26-27: "However, even at water depths deeper than 1500m 10–30% of the coastal value are reaching 10% of the grid cells at the surface (Figs. 5 and 4c, f)". Please improve this sentence.

* Page 15706. Do you really need subsection "3.2.1 Comparison of simulated and observed surface concentrations"? There is not any other subsection under 3.2.

* Page 15706, lines 11-17 (beginning of section on evaluation for the Atlantic): "Data from three cruises allow to evaluate the latitudinal gradient in the Atlantic: the Polarstern cruise Blast 2 (Butler et al., 2007), the Polarstern cruise ANT X/1 (Schall et al., 1997) which both cross the Atlantic from the Northeast (off the European and North African continents) to South America 15 in boreal fall (October, November), and the Polarstern cruise ANT XVII/1 (Chuck et al., 2005) which lead off the African coast from the subtropical North to the South Atlantic in August". This sentence is too long and needs splitting. In addition, please indicate the corresponding figures from the supplementary material to make things easy for the reader: Blast 2 (Fig. S6), ANT X/1 (Fig. S14), ANT XVII/1 (Fig. S24).

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* 15707, lines 7-10: "We simulate a global net primary productivity (NPP) of 59.3 GtCyr⁻¹, which is in the range of published estimates (e.g. 52 GtCyr⁻¹, Westberry et al., 2008, < 40 GtCyr⁻¹ >= 60 GtCyr⁻¹, mean 51 GtCyr⁻¹: between less than 40 GtCyr⁻¹ and more than 60GtCyr⁻¹, mean 51 GtCyr⁻¹, Carr et al., 2006)". This does not look right, please re-write.

* Beginning of page 15708. The following lines do not read very well: "this method ((fractional) reduction of the production ratio) does not improve uniformly the model results. Thus both, primary productivity, production rate (and species composition) need to reflect the conditions during the cruise to ...". You could change it to something like "this method (reduction of the production ratio) does not improve uniformly the model results. Ideally primary productivity, production rate and even species composition would need to reflect the conditions during the cruise ..."

* Section 3.2. Evaluation for the Pacific Ocean. I suggest changing the first sentence to something like: "To evaluate bromoform in the Pacific we look at data from four cruises in the Eastern Pacific (Blast 1, Fig. S26; Gas Ex 98, Fig. S28; Phase 1-04, Fig. S30; RB-99-06, Fig. S32; Butler et al., 2007) and one cruise in the Western Pacific (TransBrom, Fig. S34, Ziska et al., 2013); there is overlap for them". As mentioned above, indicating the figures from the very beginning makes the rest of the section easier to follow.

* page 15708, Line 11: Not sure if you need to change "spring" to "spring-summer" (see Fig. S28)

* page 15708, Line 11: "For both the northern and the equatorial east Pacific bromoform concentrations in fall and winter match observations well (Blast 1, Fig. S26 and Gas Ex 98, Fig. S28)". I think you need to change "Gas Ex 98, Fig. S28" to "RB-99-06, Fig. S32".

* Section 3.2. Evaluation for the Southern Ocean and Arctic. Page 15709, lines 4-6. Again indicate the figure numbers from the very beginning: "The comparison of

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HAMOCC simulated primary production ... along several ship tracks: ADOX (Fig. S45), CLIVAR01 (Fig. S41), SWEDARP (Fig. S37)"

* page 15709, line 12: "conclude ... to the quality" -> "conclude ... about the quality"

* page 15709, line 16: "SWEDARP (S38, Abrahamsson et al., 2004)" -> "SWEDARP (Fig. S36, Abrahamsson et al., 2004)"

* page 15709, line 20: "BLAST3 (February–April) and CLIVAR01 (October–November)". Please mention the figures: Fig. S38-S39 for BLAST3 and Fig. S40-S41 for CLIVAR01.

* page 15710, lines 20-23 (beginning of section 3.3, Gas-exchange with the atmosphere): "High emissions ... in boreal winter (DJF) in the Southern Ocean (Fig. 7b and e), in boreal summer (JJA) in the Northern Pacific and the Atlantic Oceans (Fig. 7a and d)". I think that the first reference here should be to Fig. 7a and the second one to Fig. 7e.

* page 15711, lines 4-5: "The reversal of gas-exchange depending on season implies, that ...". The comma in this sentence is not needed.

* Page 15711, lines 11-12: "Zonal maxima are higher than 0.8e-13 kg m-2 s-1 in the southern extratropics and 0.4e-13 kg m-2 s-1 in the Tropics". This does not look right since I do not see any value higher than 0.40e-13 kg m-2 s-1 in Fig. 8.

* You start "Tropics" with capital in some parts of the text. It should be lower case.

* page 15712, lines 15-16: "Therefore, we will focus in the comparison of our results with previous estimates on this inventory" -> "Therefore, we will focus on the comparison of our results with those of that inventory"

* Last lines of page 15712: "Another reason why our global emissions are lower than the ones in Ziska et al. (2013), is that their high emissions often occur in locations where no data exist, where nevertheless higher emissions are calculated based on the

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extrapolation method" → "Another reason why our global emissions are lower than the ones in Ziska et al. (2013) is that their high emissions often occur in locations where no data exist as a result from the extrapolation method used"

* page 15713, lines 10-11: "data from the Blast 2 cruise (Fig. S1, Butler et al., 2007), or data from the M60 cruise (Fig. S5, Ziska et al., 2013)". Blast 2 should be Fig. S6 and M60 is Fig. S18.

* What is the long list of numbers at the end of some references in the reference list? See e.g. Hossaini et al. (page 15717), Quack and Wallace (page 15719) or Ziska et al. (page 15721)

* Tables 2 & 3: I assume that "Clim-at" is the same as the "Ref" simulation, but this should be changed to "Ref" for consistency with the rest of the manuscript.

* Captions of Figures 1, 2, 4: "boreal summer (a) and boreal winter (d)". It should be the other way round.

* Caption of Figure 2: "Contour lines show the fraction of diatoms (b, e) and nondiatoms (c, f) in bulk phytoplankton (0.5, 0.75, 1.0 contour lines), whereby fractions > 0.5 are indicated by a mesh pattern (inclined mesh for diatoms, straight mesh for non-diatoms)". It is clear what the mesh pattern represents, but I cannot distinguish any "0.5, 0.75, 1.0 contour lines". Is that because the plots are too small?

* Figure 4: Need to write "(c, f)" before "in the same season"

* Figure 5: Is it possible to indicate how the geographical areas (i.e. Atlantic, Arctic, Pacific and Southern Ocean) have been defined? Do they correspond to some specific lat/lon intervals or do they cover the whole extension of each ocean?

* Figure 7: "boreal summer (a) and boreal winter (e)". It should be the other way round.

* Figure 8: Add "(d)" after "and Dia".

* Figure S1 in supplement: Again I guess that "Clim-at" should be changed to "Ref" for

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consistency with the rest of the paper

* Figure S3, S7, S9, ..., S53: In "mgC m⁻² dy⁻¹" change dy⁻¹ to day⁻¹ (or d⁻¹). Change "data 1997-2009" to "1997-2009 data". Change "NPP produced" to "NPP product" (or "NPP produced").

* Figure S34: Oktober → October

Please read carefully the whole text in the main manuscript. I might have missed other errors. This is still a nice manuscript, but it is a pity that because of so many technical inaccuracies one cannot focus on the science.

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