Reply to Carlos Ordóñez (referee #2)

We thank the reviewer for the thorough and helpful review. Below you find the detailed answers (normal font) to the issues raised in the review (`typewriter`). Added text blocks for the revised version of the manuscript are written in italics.

> 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 Ziskas 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 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 Ziskas 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).

We will revise the order of the paragraphs and swap the sections ”2.3 Observations” and ”2.2 Model setup”. Additionally, we will add the following sentences to ”2.2. Observa-

tions":

Gridded atmospheric mixing ratios are used as boundary conditions for the model after conservative spatial interpolation onto the model grid. Ziska et al. provide gridded data derived from a robust fit method (RF) and ordinary least square (OLS) regression. The largest difference in these methods is in the treatment of outliers. We chose to use the data derived from the RF method that is less sensitive to outliers.

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 Ziskas dataset in another question below.

The extrapolated global fields derived by Ziska et al. 2013 do not resolve the seasonal cycle of atmospheric and surface ocean concentrations as the temporal data coverage is too low. We will revise this paragraph:

The additional experiment, Seas-at, differs from Ref only by the atmospheric boundary conditions for bromoform gas-exchange. *In Seas-at atmospheric boundary conditions follow a seasonal cycle. We derive that seasonal cycle from the surface ocean concentrations calculated in experiment Ref, because the extrapolated fields of Ziska et al. 2013 do not resolve temporal variability.* In particular, the ratio between seawater concentration monthly means and their annual mean was used to construct the monthly means of atmospheric concentrations from the climatological mean.

(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?

The poorly explained numbers in brackets in Table 2 show the residence times with regard to gas-exchange and degradation. To make this more clear, Table 2 will be revised and we

will add degradation:

Table 1: Simulated global annual bromoform production and loss (Gmol $CHBr_3\,yr^{-1}$), inventory (Gmol $CHBr_3$) and residence time (days); the first number refers to gas exchange and the second number to degradation

Process	*Ref*	Seas-at	Half	Dia	NDia
Uptake	0.018	0.016	0.024	0.022	0.019
Outgassing	0.3142	0.311	0.149	0.22	0.24
Planktonic source	0.37	0.37	0.18	0.26	0.29
Degradation	0.069	0.066	0.057	0.063	0.063
Inventory	0.215	0.205	0.1822	0.1966	0.200
Residence times $\tau^{1)}$ (days)	205	197	322	253	241
($\tau^{2)}_{gasx}$, $\tau^{3)}_{degr}$) (days)	(249, 1141)	(239, 1124)	(445, 1167)	(326, 1144)	(304, 1161)

1) $\tau = \frac{1}{\frac{1}{\tau_{degr}}+\frac{1}{\tau_{gasx}}}$

2) $\tau_{gasx}=\frac{inventory}{outgassing}$

3) $\tau_{degr}=\frac{inventory}{degradation}$

(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 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?

We agree and will revise the paragraph:

The best match with observations is achieved when either reducing the bulk bromoform production rate, *or considering different production rates for different phytoplankton groups (Fig.6, S4-5).* A reduced diatom - bromoform production ratio slightly improves the representation of the bromoform concentrations in the southern hemisphere while the concentrations in the northern hemisphere are better depicted for a reduced non-diatom bromoform production ratio.

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.

We will include a heading to this paragraph: "Summary"

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 iodocarbons. The formulation of the fluxes between 20 N and 20 S is: E = 1.127e105 f r 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.
  It is true we so far omitted this feature in the discussion of the results. (see below)

- 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.

  Ziska et al. (2013) calculate their emissions from constant fields of atmospheric and oceanic concentrations. The seasonal variability in their emissions arises from the seasonality of the meteorological parameters used in the diagnosis of the gas-flux. In Hossaini et al. (2013) the climatological (time-invariant) field of emissions was used.

We will improve the discussion of the seasonality in the revised version of the manuscript:

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) *based on the OLS method. The only inventory in Hossaini et al.'s study that considers temporal variability in the emissions is the one by Ordóñez et al 2012. They indirectly resolve seasonally varying bromoform fluxes within the tropics (*$\pm 20^{\circ}$*), because they relate air-sea fluxes with satellite-based chlorophyll concentrations which are in turn temporally variable. Hossaini et al. 2013* are *however* able to reproduce most of the seasonality of bromoform atmospheric mixing ratios *even* with the temporally invariant emissions. *They argue that this is* presumably because *the seasonality* is driven by photolytic degradation in air. The seasonality in our simulated emissions sometimes encompasses a variation of more than a factor of 2*, in particular in the productive extratropical regions.* The impact of this seasonality onto the evolution of atmospheric mixing ratios needs to be tested in dynamic ocean–atmosphere coupling.

(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 Ziskas paper which might contain some errors). I believe 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.
- Ordonez 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 havent tried to do the conversion, but you give a single value without explaining why.

- Your references to Ziskas 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 dont 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.

The global flux is 0.3 Gmol $CHBr_3$ yr^{-1} as indicated in the text and shown indirectly as outgassing and uptake in Table 2. The reviewer was correct; the unit of Table 3 is Gmol Br yr^{-1}. We accidentally forgot to convert the unit when transferring the data to this table. We will update the table with the corrected value of 0.9 Gmol Br yr^{-1}. Furthermore we will update the table and follow the reviewer's suggestions concerning the fluxes in the main text:

Also at the global scale the open ocean is a bromoform source to the atmosphere, and delivers approximately 0.9 *Gmol Br* yr^{-1} (Table 3).

Previous estimates of global annual marine bromoform emissions range from *1.5-22 Gmol Br* $yr-1$ (Table 3), considering either both coastal and open ocean regions or treating them individually.

Table 3: *Previously reported and simulated* global annual bromoform net emissions ($\mathrm{Gmol\,Br\,yr^{-1}}$) from the ocean.

Source type	Lit. value	Reference
Open ocean	10.01 (3–22)	Quack and Wallace (2003)
	10.26	Yokouchi et al. (2005)
	4.75-7.06	Warwick et al. (2006)
Global ocean	10.0	Butler et al. (2007)
Open ocean	1.9	Butler et al. (2007)
	10.3	O'Brien et al. (2009)
Tropics	4.35	Palmer and Reason (2009)
Global ocean	5.31	Liang et al. (2010)
Open ocean	3.19	Liang et al. (2010)
	6.33	Ordóñez et al. (2012)
Global ocean	2.49	Ziska et al. (2013) (OLS)
Global ocean	1.5	Ziska et al. (2013) (RF)
Global ocean	3.5	Sousa Santos and Rast (2013), Sousa Santos (2009)
Open ocean	0.9	This study: Ref, Seas-at (net flux)

(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; Ordez et al., 2012). The authors should discuss whether potential issues in the model (e.g. missing bromoform 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.

We will include a discussion to the section "Gas exchange" and a statement in the section "Conclusions":

3.3 Gas exchange with the atmosphere:
Also at the global scale the open ocean is a bromoform source to the atmosphere, and delivers approximately 0.3 Gmol $CHBr_3$ yr^{-1} (Table 3). *For the Northern Atlantic and the Arctic Ocean, the experiment Coast suggests that coastal sources could enhance oceanic concentrations and counteract the undersaturation of the ocean. Furthermore, in the Arctic and Southern Ocean bromoform production in sea ice could have a similar effect with an increase sea-air flux also not resolved in the model. Both mechanisms are currently not included but would lead to higher simulated global bromoform emissions. In addition the seasonal reversal of gas-exchange is also strongly influenced by the atmospheric boundary conditions. Thus, it is important to choose these carefully for simulating realistic bromoform emissions with a stand-alone ocean model.*

4. Conclusions:
Particularly interesting are the large-scale patterns that indicate bromoform uptake from the atmosphere, which do not show up in climatological mean emissions. *The robustness and implications of the flux reversal should be studied in more detail. In this regard the model needs to be refined to resolve also coastal sources and bromoform production within sea ice.*

(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!

We improved the artwork. Below you find the updated versions of figures 1, 2, 4, 7, S1, and S3.

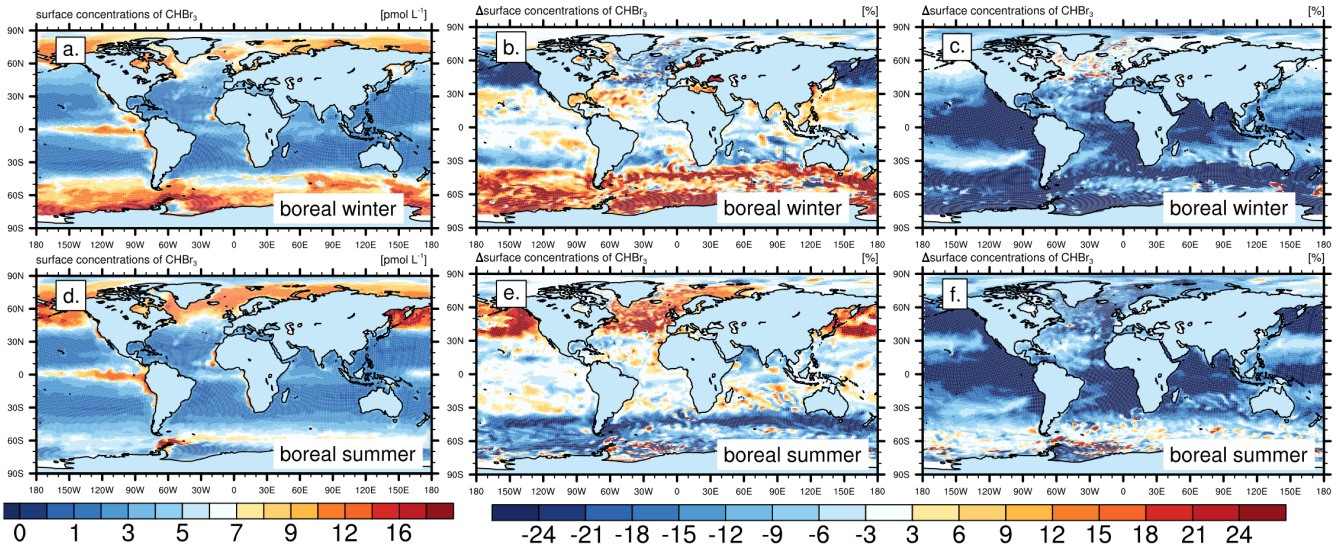


Figure 1: Mean surface bromoform concentrations ($\mathrm{pmol\,L^{-1}}$) in experiment *Ref* in boreal winter **(a)** and boreal summer **(d)**, percentage difference (e.g. $100 \cdot \frac{Seas-at-Ref}{Ref}$) of *Seas-at* **(b, e)** and *Half* **(c, f)** in the same season.

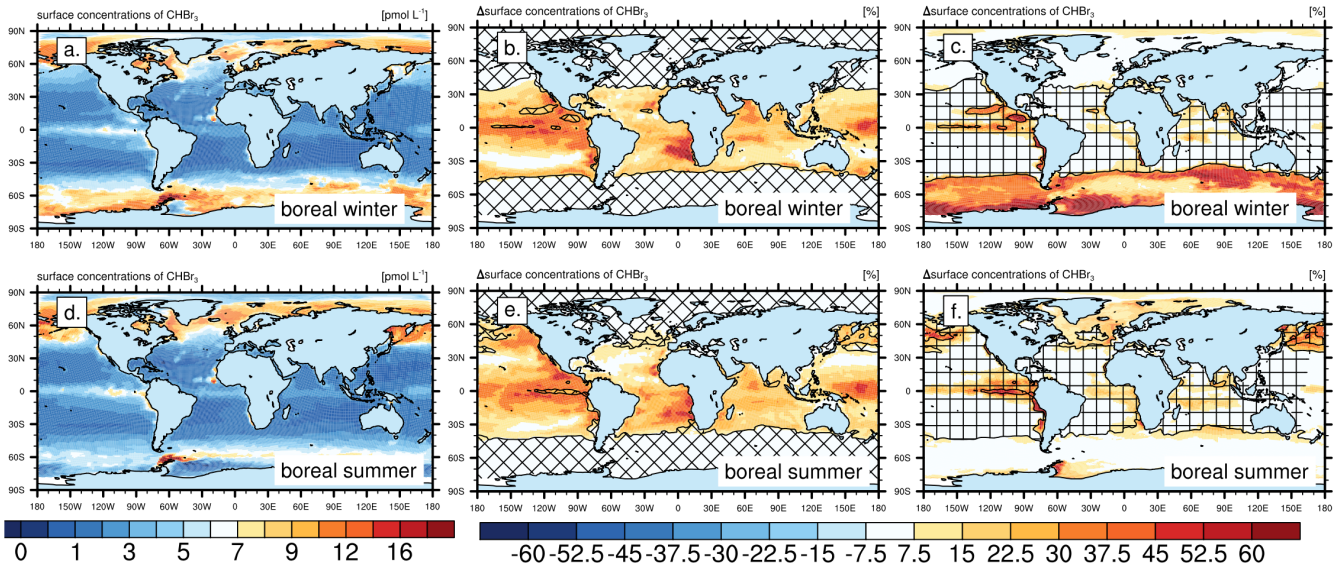


Figure 2: Mean surface bromoform concentrations (pmol L^{-1}) in experiment *Half* in boreal winter **(a)** and boreal summer **(d)**, percentage difference (e.g. $100 \cdot \frac{Dia-Half}{Half}$) of *Dia* **(b, e)** and *NDia* **(c, f)** in the same season. Mesh patterns show regions where the fraction of diatoms **(b, e)** or non-diatoms **(c, f)** in bulk phytoplankton dominates (i.e. fraction > 0.5) (inclined mesh for diatoms, straight mesh for non-diatoms).

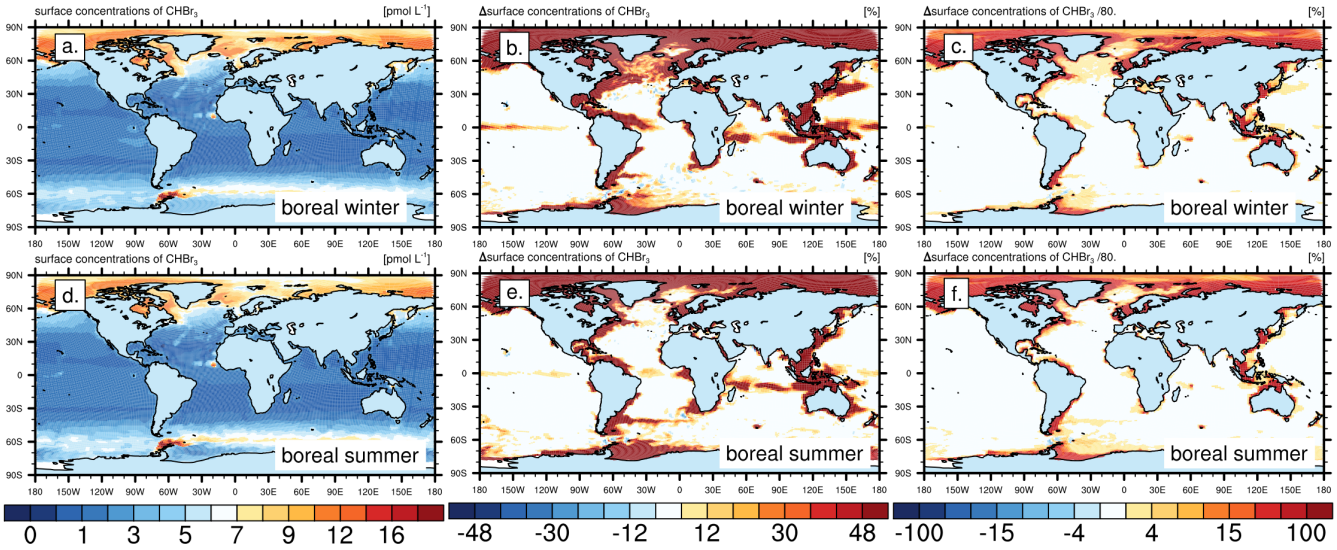


Figure 4: Mean surface bromoform concentrations ($\mathrm{pmol\,L^{-1}}$) in experiment *Equi* in boreal winter **(a)** and boreal summer **(d)**, percentage difference (e.g. $100 \cdot \frac{Coast-Equi}{Equi}$) of experiment *Coast* **(b, e)** and $100 \cdot \frac{Coast-Equi}{80\,\mathrm{pmol\,L^{-1}}}$ **(c, f)** in the same season.

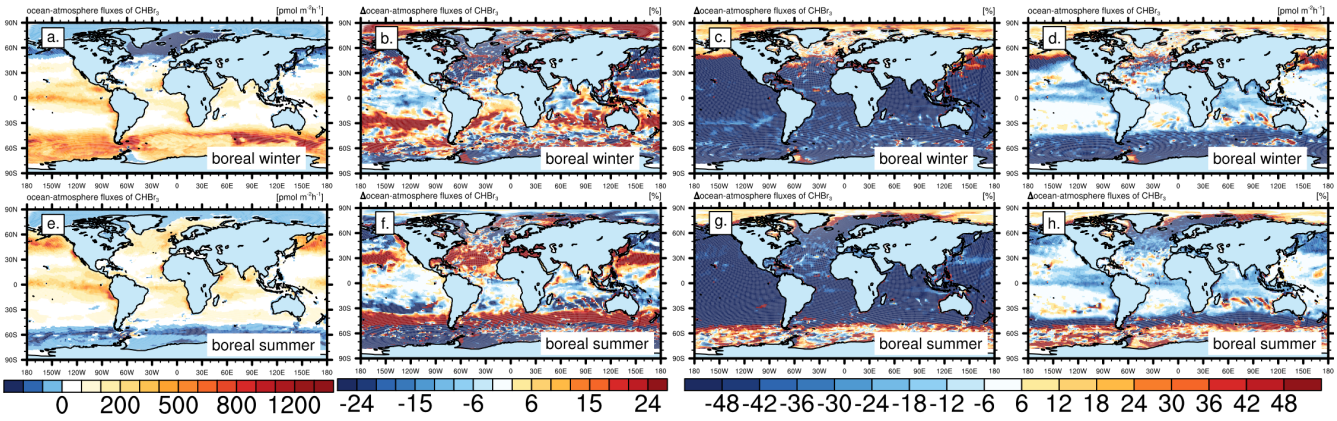


Figure 7: Mean bromoform sea–air flux ($\mathrm{pmol\,m^{-2}\,h^{-1}}$) in experiment *Ref* in boreal winter **(a)** and boreal summer **(e)**, percentage difference (e.g. $100 \cdot \frac{Seas-at-Ref}{Ref}$) of *Seas-at* **(b, f)**, *Half* **(c, g)**, and *Dia* **(d, h)** in the same season.

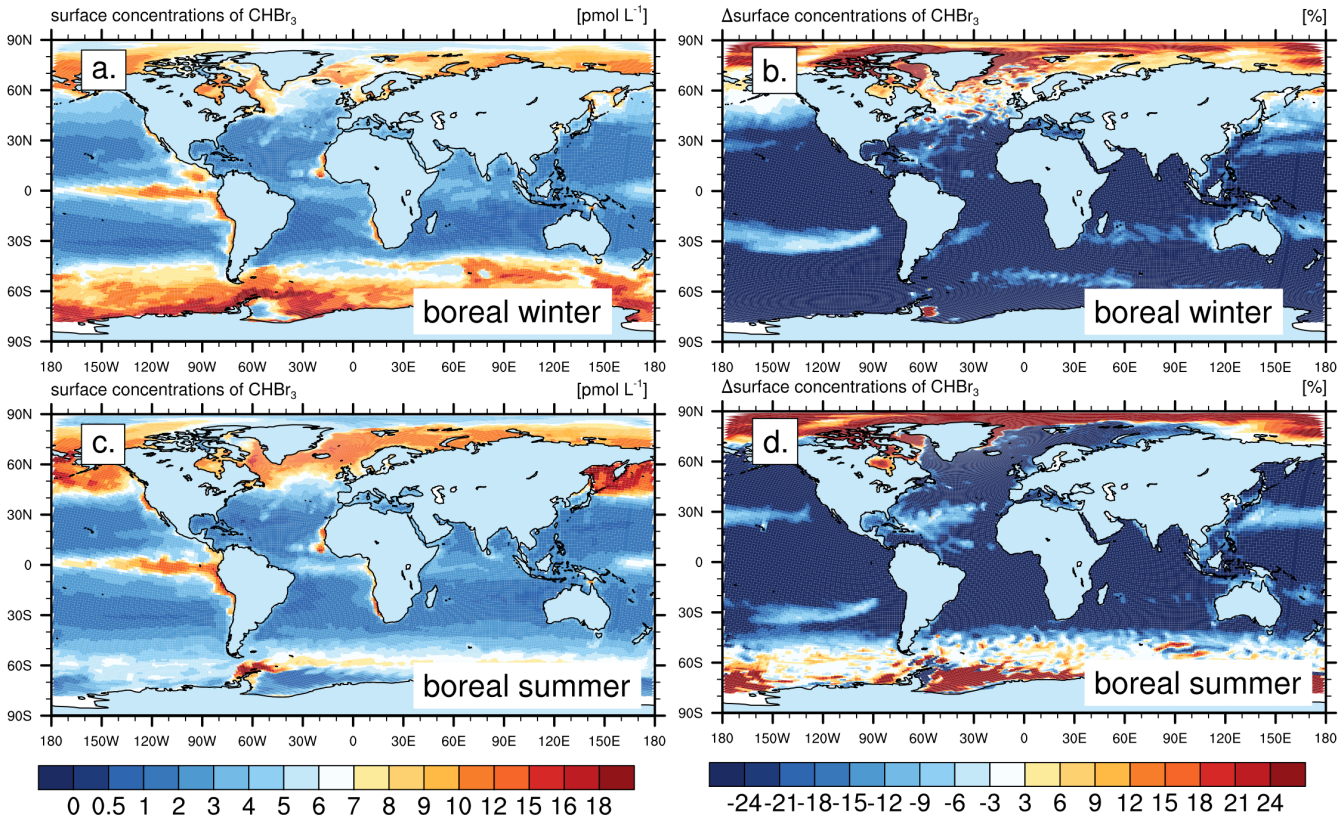


Figure S1: Bromform surface concentrations (pmol L^{-1}) in boreal winter and summer in experiment *Ref* **(a, c)** and percentage differences to concentrations in experiment *Equi* ($100 \cdot \frac{Equi-Ref}{Ref}$) **(b, d)**.

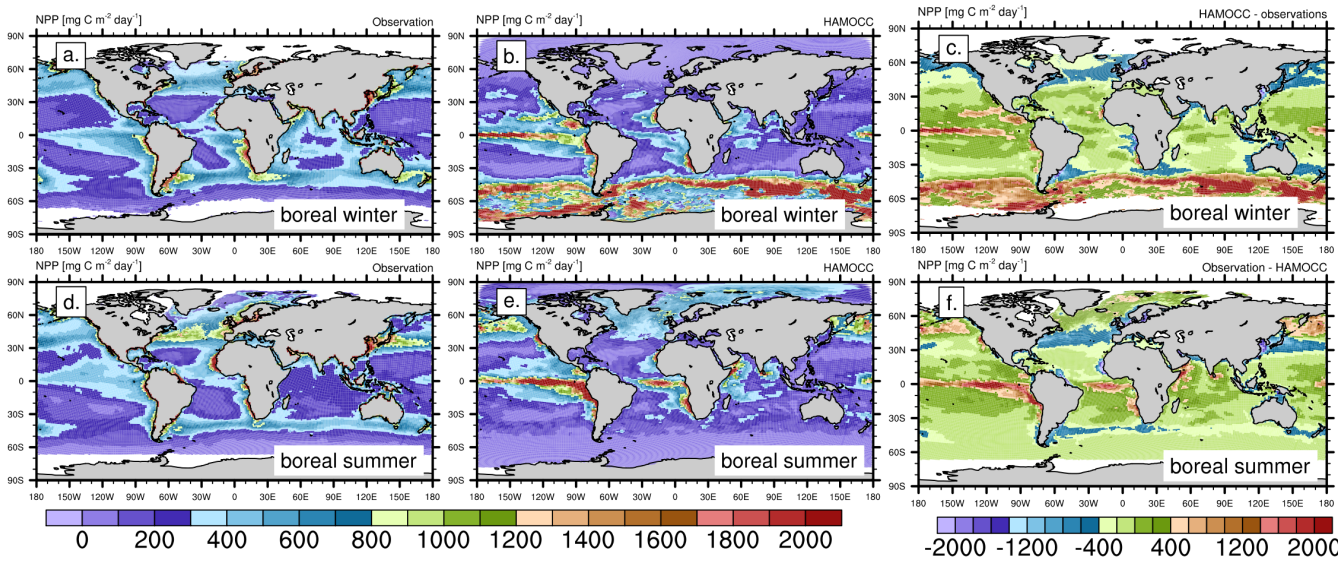


Figure S3: Simulated **(b, e)** and observation-based **(a, d)** net primary productivity ($\mathrm{mg\,C\,m^{-2}\,day^{-1}}$) and their difference **(c, f)**. The observation-based NPP product is based on data 1997-2009 from SeaWiFS Chl-*a*, PAR and AVHRR SST and derived using the VGPM model (Behrenfeld and Falkowski, 1997). The NPP product was downloaded from http://wiki.icess.ucsb.edu/measures/NPP_Products (accessed June 2014).

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 ...’’.

We will rephrase the sentence including additionally the suggestion by C.Palmer:

Bromoform ($CHBr_3$) is one of the most abundant bromine containing volatile *halocarbons* and is *a* considerable source *of* reactive bromine species in the atmosphere.

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.

We will include the reference. You find the full citation in the reference list at the end of this document.

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

We will correct the sentence:

As the biogeochemistry model does not resolve plankton functional groups, we can not directly calculate species- (or group-) specific bromoform production.

Ideally all equations should be followed by an identifier, i.e. (1), (2), ..., but I dont know if that is required by this journal.
We will follow this suggestion.

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 ...’’

The punctuation will be corrected:

We omit degradation during remineralization of detritus in this study, because Hense and Quack (2009) show that it leads to unrealistic accumulation of bromoform in the deep ocean.

As it was shown for freshwater nitrifiers that these bacteria can oxidize volatile halogenated organic compounds (including $CHBr_3$, see e.g. Sayavedra-Soto et al. (2010)) during oxidation of ammonium hydroxylamine it seems reasonable to exclude this process for low oxygen conditions.

As both processes are strongly temperature dependent and follow different kinetics, hydrolysis and halogen substitution are implemented as separate sinks in the current study.

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

We will follow this suggestion:

We modified the description of the transfer velocity given by Nightingale et al. (2000) to resolve the temperature dependence of the Schmidt number $Sc_{\mathrm{CHBr_3}}$ (Quack and Wallace, 2003):

$$k_{\mathrm{w}} = (0.222u^2 + 0.33u) \cdot \sqrt{\frac{660}{Sc_{\mathrm{CHBr_3}}}} \tag{7}$$

$$Sc_{\mathrm{CHBr_3}} = 4662.8 - 319.45T + 9.9012T^2 - 0.1159T^3 \tag{8}$$

In the equations u denotes wind speed ($m\,s^{-1}$) and T temperature (K).

page 15701. When describing the model simulations you wrote ‘‘preindustrial conditions (pCO2 = 278 ppm)’’ and ‘‘present-day conditions (pCO2 = 353 ppm)’’. pCO2 should refer to a partial pressure of CO2 in the atmosphere while you are giving values of atmospheric mixing ratios. Please re-write.

We will correct the sentence:
For all simulations the model *restarts* from a 1000 year spin up under preindustrial conditions (CO_2 = 278 ppm) followed by a 200 year spin-up under present-day conditions (CO_2 = 353 ppm).

page 15702, line 15: Change ‘‘Chl a’’ to ‘‘Chl-a’’
We will correct the sentence:

It was calculated from NASA's SeaWIFS (Sea-viewing Wide Field-of-view Sensor) level 3 data (PAR and Chl-*a*) and NOAA's AVHRR (Advanced Very High Resolution Radiometer) sea surface temperature for 1997–2009 using the Vertically Generalized Productivity Model (VGPM) (Behrenfeld and Falkowski, 1997).

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.

We will correct the sentence:

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 *winter* seasons, Fig. 1c and f).

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 CHBr3 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.

We will rephrase the sentence:

Similarly, differences between *Ref* and *Seas-at* are highest where the seasonal cycle of $CHBr_3$ surface concentrations is strong because of the variability of bromoform production (e.g. in the extratropics (Figs.1 a,b,d,e around 50° N)). This is because the seasonal cycle of the atmospheric forcing field in *Seas-at* is derived from the sea surface concentrations.

page 15704, line 3: ‘‘As mentioned above, bromoform distribution patterns for the main part follow ...’’. What do you mean by "for the main part"
"For the main part" is used to express that bromform distribution patterns almost everywhere follow production. However, there are regions where bromoform concentration patterns deviate from production patterns. We will replace "for the main part" by *mainly*.

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.

No, in this context we only refer to 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 CHBr3 inventory is similar to the impact of reducing the bulk production rate by the same factor (Table 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.

We will rephrase the sentence:
Similar to experiment Half, the reduction of the production rate in Dia leads to a reduction of the global bromoform inventory, as diatoms dominate in productive regions.

page 15704, line 19: focusing onto > focusing on
Will be corrected:
When focusing *on* certain regions though, differences in the two approaches become ap-

parent, e.g. in lower latitudes where non-diatom species dominate and the bromoform production (and concentration) is hence higher in *Dia* than in *Half*.

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 (mediterranian 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)’’.

Here "mediterranean" is used to describe "surrounded by land masses" in contrast to "the Mediterranean Sea". We will rephrase the sentence:
As expected, lateral transport from shelf regions is particularly relevant in the Arctic surface ocean (Figs. 4c, f and 5), because the Arctic Sea *is semi-enclosed by land and outgassing at cold temperatures is low.*

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.

We are going to rephrase the sentence:

However, even in deep open ocean waters (water depth > 1500m) surface bromoform concentrations reach 10-30 % of the coastal value in 10 % of the model grid cells.

Page 15706. Dou you really need subsection ‘‘3.2.1 Comparison of simulated and observed surface concentrations’’? There is not any other subsection under 3.2.

No, it is indeed redundant. We will remove this subsection heading.

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).

We will rephrase the sentence and add the figure references:
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), and the Polarstern cruise ANT XVII/1 Chuck et al., 2005). Blast 2 (Fig. S6) and ANT X/1 (Fig.S14) cross the Atlantic from the northeast (off the European and North African continents) to South America in boreal fall (October, November). The cruise ANT XVII/1 leads along the African coast from the subtropical North to the South Atlantic in August (Fig.S24).*

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.

We will rephrase the sentence:
We simulate a global net primary productivity (NPP) of 59.3 GtCyr^{-1}, which is in the range of published estimates (e.g. NPP=52 GtCyr^{-1}, Westberry et al., 2008, NPP=51 $\pm 10\,\mathrm{GtCyr}^{-1}$, Carr et al., 2006)

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 ...’’

We will follow the reviewer's suggestions:
The comparison between other individual ship cruises, e.g. MSM 18/3 (Fig. S12) and DRIVE (Fig. S10) shows that this method (reduction of the production ratio) does not improve uniformly the model results. *Ideally simulated* primary productivity, production rate *and even species composition* would need to reflect the conditions during the cruise to obtain the best possible representation of bromoform distribution patterns.

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.

We will follow this suggestion and add figure references to the first sentence of the paragraph:
To evaluate bromoform in the Pacific we closer look at data from four cruises in the Eastern Pacific (Blast 1, Gas Ex 98, Phase 1-04, RB-99-06 (Figs. S26-S33, Butler et al., 2007) and one cruise in the Western Pacific (TransBrom, Fig.S34-35, Ziska et al., 2013); please note that overlaps exist.

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

Yes, the cruise was from Mai to July, we will change the sentence:
In *spring-summer* concentrations in the model along the same track (Gas Ex 98) are at some locations three times higher than observations (Fig. S28), likely because primary production is overestimated by the model (Fig. S29).

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''.

We will correct the sentence:
For both the northern and the equatorial east Pacific bromoform concentrations in fall and winter match observations well (Blast 1, Fig. S26 and *RB-99-06, Fig. S32*).

Section 3.2. Evaluation for the Southerh Ocean and Arctic. Page 15709, lines 4- 6. Again indicate the figure numbers from the very beginning: ''The comparison of HAMOCC simulated primary production ... along several ship tracks: ADOX (Fig. S45), CLIVAR01 (Fig. S41), SWEDARP (Fig. S37)''

We will follow this suggestion and add figure references:
The comparison of HAMOCC simulated primary production to the one derived by the VGPM model shows that NPP is overestimated in austral summer (Fig. S3) and along several ship tracks *(ADOX, Fig.S44-45, CLIVAR01, Fig.S40-41 SWEDARP, Fig. S36-37)*.

page 15709, line 12: ''conclude ... to the quality'' > ''conclude ... about the quality''
We will revise the sentence:
For the Southern Ocean it is difficult to directly conclude from deviations between simulated and observed NPP *about* the quality of simulated bromoform.

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

We will correct the figure reference:
This can be also seen for SWEDARP (Fig.*S36*, Abrahamsson et al, 2004), where bromoform concentrations do not follow the pattern of primary productivity or chlorophyll in both model results and observations.

page 15709, line 20: ‘‘BLAST3 (FebruaryApril) and CLIVAR01 (October November)’’. Please mention the figures: Fig. S38-S39 for BLAST3 and Fig. S40-S41 for CLIVAR01.
We will include the figure references:
However, there are also examples for a good model representation of observed bromoform concentrations and primary production, i.e. for BLAST3 (February–April, *Figs.S38-39*) and CLIVAR01 (October–November, *Figs.S40-41*) (140–250° E).

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.
We will correct the sentence:
High emissions ($> 1200\,\mathrm{pmol\,m^{-2}\,h^{-1}}$) occur in regions of high bromoform production, i.e. in boreal winter (DJF) in the Southern Ocean (Fig. 7*a*), in boreal summer (JJA) in the Northern Pacific and the Atlantic Oceans (Fig. 7*b*).

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

We will rephrase the sentence (see above, comment on flux reversal):
Thus, it is important to choose these carefully for simulating realistic bromoform emissions with a stand-alone ocean model.

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.

Fig8. shows median values and not zonal maxima. We will include a "not shown" to make this more clear.:

Zonal maxima are higher than $0.8\times10^{-13}\,\mathrm{kg\,m^{-2}\,s^{-1}}$ in the southern extratropics *compared to* $0.4\times10^{-13}\,\mathrm{kg\,m^{-2}\,s^{-1}}$ in the *tropics* (not shown), which is in contrast to the often

assumed distribution that shows largest emissions from the tropical oceans (Quack et al., 2004, Warwick et al. 2006, Sousa Santos & Rast, 2013).

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

We will correct this. See above and:
The approaches differ in the number and extent of these zones, the treatment of the *t*ropics and coastal regions, and the temporal resolution considered.

In Hossaini et al.(2013) good agreement between observed and simulated atmospheric mixing ratios, in particular within the *t*ropics could be achieved when using the emission inventory by Ziska et al. (2013), which was the lowest of the previous estimates (Table T2).

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’’

We will rephrase the sentence:
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 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’’
We will rephrase the sentence:
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*, e.g. in the northern North Atlantic and in the subtropical East South Pacific.

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))’’. Bast 2 should be Fig. S6 and M60 is Fig. S18.
The figure references will be corrected:
Here, i.e. in the subtropical Atlantic, modelled concentrations match observations well (e.g. compared to data from the Blast 2 cruise (Fig. *S6* Butler.King.ea2007a), or data from the M60 cruise (Fig. *S18*, Ziska et al., 2013).

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)

These numbers were included by the Copernicus typesetting team and indicate the pages where the references occur.

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.

The table will be corrected:

Table 2: Simulated global annual bromoform production and loss (Gmol $CHBr_3\,yr^{-1}$), inventory (Gmol $CHBr_3$) and residence time (days); the first number refers to gas exchange and the second number to degradation.

Process	*Ref*	Seas-at	Half	Dia	NDia
Uptake	0.018	0.016	0.024	0.022	0.019
Outgassing	0.3142	0.311	0.149	0.22	0.24
Planktonic source	0.37	0.37	0.18	0.26	0.29
Degradation	0.069	0.066	0.057	0.063	0.063
Inventory	0.215	0.205	0.1822	0.1966	0.200
Residence times $\tau^{1)}$ (days)	205	197	322	253	241
($\tau^{2)}_{gasx}$, $\tau^{3)}_{degr}$) (days)	(249, 1141)	(239, 1124)	(445, 1167)	(326, 1144)	(304, 1161)

1) $\tau = \frac{1}{\frac{1}{\tau_{degr}}+\frac{1}{\tau_{gasx}}}$

2) $\tau_{gasx}=\frac{inventory}{outgassing}$

3) $\tau_{degr}=\frac{inventory}{degradation}$

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

The captions will be corrected for the revised versions of the figures (see above).

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?

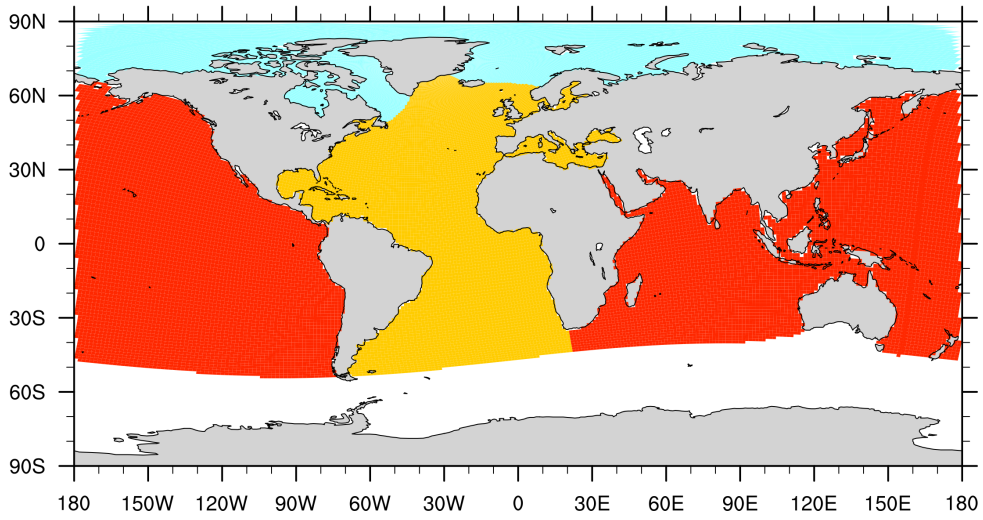
These contour lines where removed in the revised version of the figure (see above).

Figure 4: Need to write ‘‘(c, f)’’ before ‘‘in the same season’’
The caption will be corrected :

Mean surface bromoform concentrations (pmol L^{-1}) in experiment *Equi* in boreal winter **(a)** and boreal summer **(d)**, percentage difference (e.g. $100 \cdot \frac{Coast-Equi}{Equi}$) of experiment *Coast* **(b, e)** and $100 \cdot \frac{Coast-Equi}{80\,\text{pmol}\,L^{-1}}$ **(c, f)** 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?`

The extent of the ocean basins is shown below.



We will not show this figure in the manuscript. Fig.5 was updated including subplot labels a-d and a corrected naming of the histograms for the Pacific **and Indian Ocean** (see below).

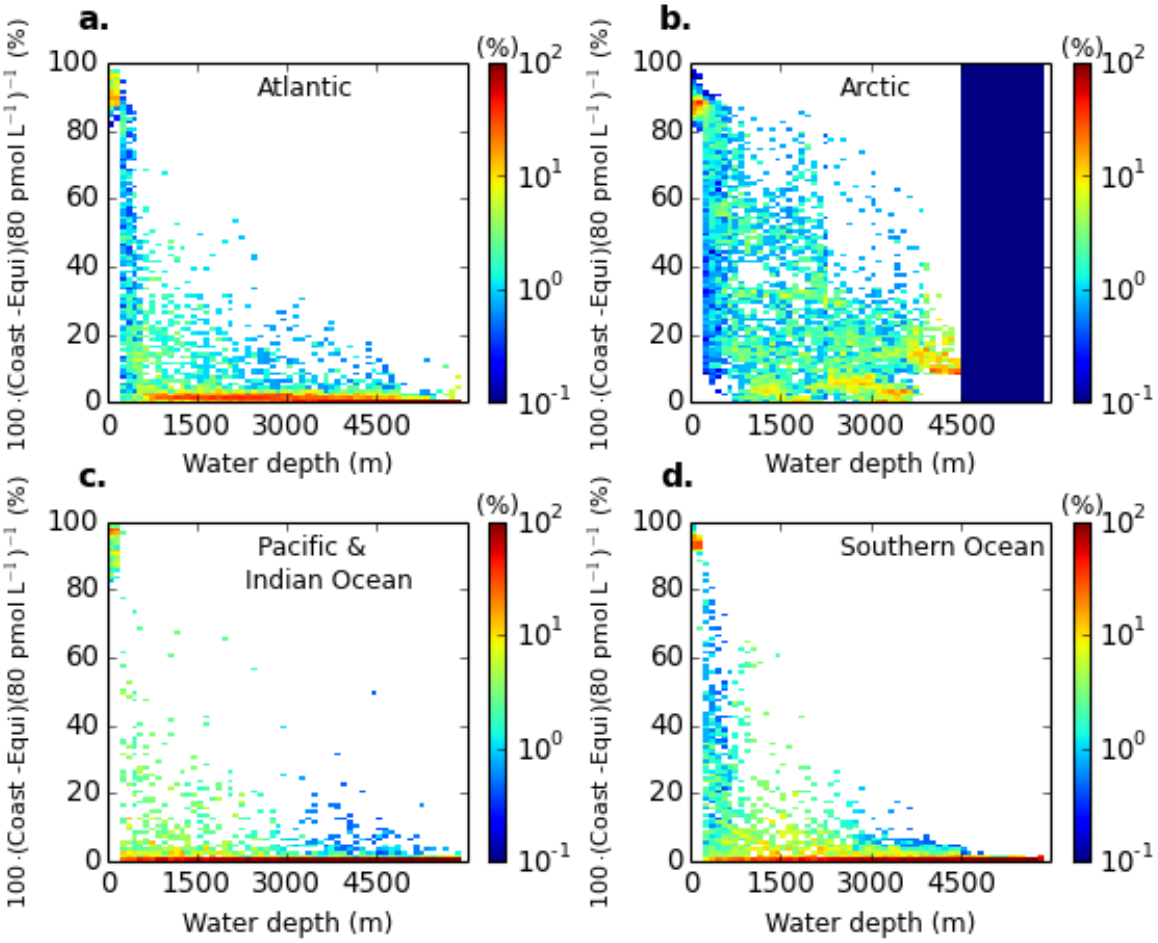


Figure S5: Histograms of $100 \cdot \frac{Coast-Equi}{80\,\mathrm{pmol\,L^{-1}}}$ surface concentrations [%] for different local water depths in the Atlantic Ocean (**a**), Arctic Ocean (**b**), Pacific and Indian Ocean (**c**), and Southern Ocean (**d**).

`Figure 7: ‘‘boreal summer (a) and boreal winter (e)’’. It should be the other way round.`
The figure caption will be corrected:
Mean bromoform sea–air flux ($\mathrm{pmol\,m^{-2}\,h^{-1}}$) in experiment *Ref* in *boreal winter* **(a)** and *boreal summer* **(e)**, percentage difference (e.g. $100 \cdot \frac{Seas-at-Ref}{Ref}$) of *Seas-at* **(b, f)**, *Half* **(c, g)**, and *Dia* **(d, h)** in the same season.

`Figure 8: Add ‘‘(d)’’ after ‘‘and Dia’’.`
The figure caption will be revised:

Zonal median of bromoform sea–air flux ($\mathrm{kg\,m^{-2}\,s^{-1}}$), mean of JJA (blue), DJF (black), MAM (green), and SON (orange), and annual mean (dashed gray). Results are from *Ref* **(a)**, *Seas-at* **(b)**, and *Half* **(c)**, and *Dia* **(d)**.

`Figure S1 in supplement: Again I guess that ‘‘Clim-at’’ should be changed to ‘‘Ref’’ for consistency with the rest of the paper.`
We will correct the figure caption:
Bromform surface concentrations ($\mathrm{pmol\,L^{-1}}$) in boreal winter and summer in experiment *Ref* (a, c) and percentage differences to concentrations in experiment Equi ($100 \cdot \frac{\mathrm{Equi-Ref}}{\mathrm{Ref}}$) (b, d).

`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 producted’’ to ‘‘NPP product’’ (or ‘‘NPP produced’’).`

We will correct the figure captions:
Simulated (b, e) and observation-based (a, d) net primary productivity ($\mathrm{mg\,C\,m^{-2}\,day^{-1}}$) and their difference (c, f). The observation-based NPP product is based on data 1997-2009 from SeaWiFS Chl-*a*, PAR and AVHRR SST and derived using the VGPM model (Behrenfeld and Falkowski, 1997). The NPP product was downloaded from `http://wiki.icess.ucsb.edu/measures/NPP_Products` (accessed June 2014).

Simulated and observation-based net primary productivity ($\mathrm{mg\,C\,m^{-2}\,day^{-1}}$). Green shades show minimum and maximum range of the observation-based estimate, the black dashed line shows the median. The observation-based NPP product is based on data 1997-2009 from SeaWiFS Chl-*a*, PAR and AVHRR SST and derived using the VGPM model (Behrenfeld and Falkowski, 1997). The NPP product was downloaded from `http://wiki.icess.ucsb.edu/measures/NPP_Products` (accessed June 2014).

`Figure S34: Oktober > October`

We will correct the figure caption:
Observations are from the R/V Sonne cruise TransBrom Sonne in *October* 2009 as listed in the SI of (Ziska et al., 2013).

`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.`

We really appreciate the helpful comments and will additionally ask colleagues to proofread the revised version.

References

Hughes, C., Johnson, M., Utting, R., Turner, S., Malin, G., Clarke, A., and Liss, P.: Microbial control of bromocarbon concentrations in coastal waters of the western Antarctic Peninsula, Marine Chemistry, 151, 35–46, 2013.

Sousa Santos, G.: The Effect of Halogens on global tropospheric Ozone, Reports on Earth System Science, 59, 2009, available at: http://www.mpimet.mpg.de/fileadmin/publikationen/Reports/WEB_BzE_59.pdf.