Reply to Carl J. Palmer

We thank the reviewer for his helpful comments and apologize for the sloppy preparation of the manuscript. 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.

`Page 15694 Line 24-25: "volatile halocarbon and is one considerable source for reactive bromine species" is ambiguous and not well written, would suggest "volatile halocarbonS and is a globally significant source OF reactive bromine species"`

We will rephrase the sentence:

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 4: The words "consequently on climate" as currently structured implies a much more significant and proven link with climate than the current data are able to substantiate. I would advise rephrasing to make it clear that this is merely a potential link.`

We will drop "consequently on climate":

Due to its life-time of approximately 3–4 weeks [..] it alters the bromine budget in both the troposphere and the stratosphere and can lead to ozone depletion with potential impacts on the radiation budget of the atmosphere [..]

`Line 11: replace "only" with "relatively"`
Will be rephrased:
Anthropogenic sources [..] are thought to contribute *relatively* little to the global emissions [..].

`Line 15:25: I would suggest that the link between bromoperoxidase activity and cell growth is overstated here and conversely discussion of the demonstrated link between this enzymes activity and oxidative stress is absent. Whereas, this omission by no means undermines the science presented (in phytoplankton the bulk emission of CHBR3 may well best correlate with the growth phase) it does however warrant discussion here. E.g. Pedersen, 1996.`

We will modify the paragraph:
Bromoform synthesis in the open ocean is mainly related to phytoplankton (Moore et al., 1996; Lin and Manley, 2012). However, it is unclear whether bromoform is formed extra-

or intracellularly. In any case, the enzyme bromoperoxydase catalyzes the process in which bromide is oxidized in the presence of H_2O_2 followed by a halogenation of organic compounds (the haloform reaction). There are indications for intracellular production, e.g. some laboratory studies show that bromoform is released during phytoplankton growth (e.g. by diatoms, Moore et al., 1996; Hughes et al., 2013). In contrast, there is also evidence that bromoform is extracellularly produced, as the components that are necessary for bromoform production (dissolved organic compounds and the enzyme bromoperoxidase) may escape via cell lysis or exudation of phytoplankton (Lin and Manley, 2012; Wever and van der Horst, 2013).
Enhanced bromoform production during stress, as shown for macroalgae (e.g. Bondu et al. (2008)), has not been demonstrated for phytoplankton. However, the amount of bromoform produced can be related to different phytoplankton species. Differences between typical open ocean microalgae, i.e. the coccolithophores (Emiliana and Calcidiscus) and diatoms (Chaetoceros) are rather small (within a factor of 2) (Colomb et al., 2008). These different phytoplankton groups show different global distribution patterns.

`Line 26: The reaction with DOM in seawater is presumably assumed to proceed via the haloform reaction and if so this should be explicitly stated.`

The paragraph on bromoform production will be modified and the haloform reaction will be mentioned (see above).

`Page 15696 Some acronyms used here are not defined.`

The acronyms will be defined:

For this purpose we implement a refined version of the bromoform module of Hense and Quack (2009) into *a marine biogeochemistry model (the Hamburg Ocean Carbon Cycle model HAMOCC: Ilyina et al., 2013) which is coupled to* a global ocean general circulation model (*the Max Planck Institute ocean model,* MPIOM: Marsland et al., 2003).

At that time the model was used within the one-dimensional water column model GOTM (*General ocean turbulence model,* Umlauf et al., 2005) together with a simple *Nutrients-Phytoplankton-Zooplankton-Detritus*(NPZD)-type ecosystem model tuned to represent conditions during the Meteor Cruise M55 in the Cape Verde region. Here, we use the module within the three-dimensional ocean general circulation model *MPIOM (Marsland et al. 2003) that includes the biogeochemistry model HAMOCC (Ilyina et al., 2013).*

`Page 15700 Line 1: To help make the experimental easier to follow propose change to "Seven model experiments were ..(table 1). Of these four .."`

We will rephrase the sentence:

Seven model experiments were designed to assess different aspects of bromoform cycling (Table 1). *Of these four* experiments were designed to study bromoform synthesis by phytoplankton.

`The tense of in the document is muddled with consecutive sentence often switching between past and present e.g. "We conducted two joint experiments. In each experiment we eliminate the ..". I am aware that it is no longer a requirement to write in the past perfect tense, but nonetheless at several points such as this, the switching makes the document unnecessarily difficult to follow.`

We will revise the document and use a consistent tense.

`Page 15703 Line 23: "However, there are some uncertainties related to the production and concentration of bromoform". This line is a truism (there are some uncertainties in all data) and in its current form adds nothing. Remove/rewrite.`

We agree and specified our statement.

In the polar regions bromoform production in the model is very low, as primary production is limited by light availability even during summer, because of the sea ice. *However, particularly in this specific region uncertainties are large and bromoform cycling is not well captured in the model.*

`Page 15704 Line 3: "As mentioned above, bromoform distribution patterns for the main part follow the patterns if primary productivity" This is rather ambiguous-do you mean in your model data or in the observational data presented here?`

As indicated by the section title, all statements refer to simulated concentrations. To make this more clear we will add "simulated" to the sentence:

As mentioned above, *simulated* bromoform distribution patterns for the main part follow the patterns of *simulated* primary productivity.

`Line 11: "due to the setup" is colloquial and rather avoided, suggest rather "As a direct consequence of the experimental parameters"`

We will rephrase the sentence:

As a direct consequence of the model configuration the bromoform production in both experiments is lower than in the experiment *Ref*, and bromoform concentrations are consequently lower.

`Page 15707 Line 19-21: In this lines it is suggested that the satellite data is not capturing the conditions experienced. I would be interested to know if there was a precedent for this (and then cite it) or if this is purely speculative (in which case either acknowledge that its purely speculative or remove entirely)`

The statement lists possible causes to explain the deviations between simulated and observed bromoform concentrations: We first look at primary production (PP) as a source of bromoform. Unfortunately, primary productivity was not measured directly during the Southern Ocean cruise. Therefore, we determine PP by using the Vertically Generalized Production Model (VGPM). VGPM derives PP from satellite-based chlorophyll, photosynthetic active radiation (PAR) and SST. However, the calculated PP can provide only a rough estimate: First, the time at which satellite data and cruise data for the same location are obtained are not necessarily the same due to the satellite's orbit. In situ chlorophyll concentrations during the cruise might be different compared to the ones derived from satellite. Second, there are uncertainties related to the input parameters in general, e.g. cholorophyll is spatially and temporally highly variable. The good match between our simulated PP and the satellite-based PP along the cruise track may therefore be just by coincidence. Thus, the deviations we see in the simulated bromoform concentration may be explained by: 1) the bromoform production mechanism is not captured adequately by the model, 2) the satellite-based PP does not reflect conditions during the cruise.

`Page 15708 Line 1: The double bracket is a little confusing, maybe a square bracket could replace one set should journal conventions allow.`

We will rephrase the sentence in response to a comment by Carlos Ordóñez (referee #2):

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* 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.

`References: Hughes 2013 paper refereed to in text is not listed in references. Figure 4: c&f are missing from caption. Figure 5: labels a-d would aid clarity.`

We will include the missing reference. Here it is listed in the reference list at the end of the document.

We will correct the figure caption:

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.

We will revise figure 5 for better readability of the axes labels and will include the labels a-d:

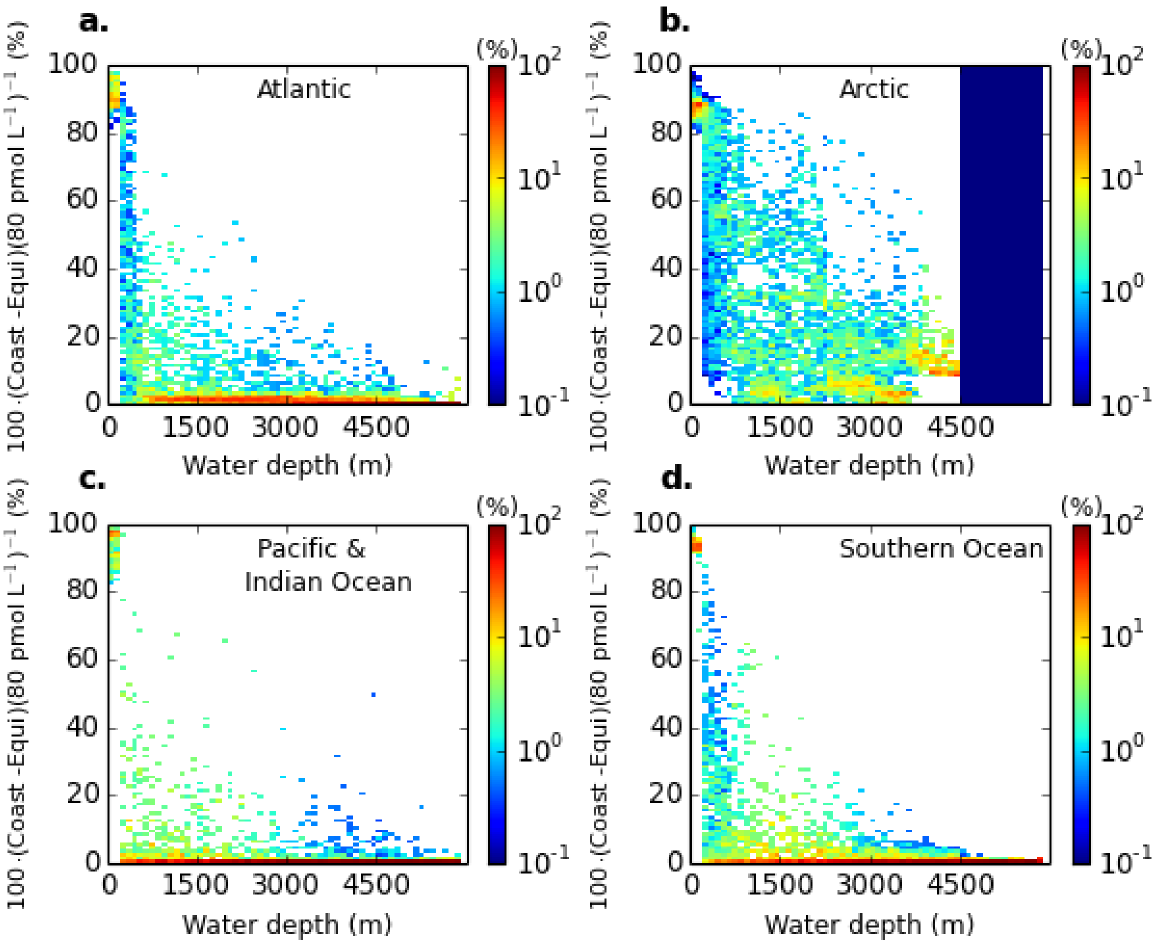

Figure 5: Histograms of $100 \cdot \frac{Coast-Equi}{80\,\mathrm{pmol\,L^{-1}}}$ surface concentrations [%] for different local water depths in the Atlantic Ocean (**a**), Arctic Ocean (**b**), Pacific and Indian Ocean (**c**), and Southern Ocean (**d**).

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.

Bondu, S., Cocquempot, B., Deslandes, E., and Morin, P.: Effects of salt and light stress on the release of volatile halogenated organic compounds by Solieria chordalis: A laboratory incubation study, Botanica Marina, 51, 485–492, 2008.