Anonymous Referee #2

***We would like to thank the reviewer for the time spent reviewing the manuscript, and for the helpful and generous comments – always greatly appreciated. Our specific responses are given below in italics*:**

[Referee] This paper describes the temporal dynamics of 11 different halocarbons during a period of 27 days in 9 CO2-manipulated mesocosms that contain polar seawater from the west coast of Spitsbergen. Over the course of the experiments, the mesocosms received additional nutrients twice. As a consequence, phytoplankton parameters show three distinct biomass peaks and hence all results are discussed within the structure of these three phases. Except maybe for M1, which appears an outlier with respect to halocarbon dynamics, all mesocosms appear to perform very similar and reproducible, which is an achievement in itself. As far as I can judge, the methods applied in this paper are sound and accurately carried out and the result is a solid database of a large range of climatically important halocarbons. The authors do a good job in organising the results of the halocarbons in 3 different groups based on their common biological production pathways and removal mechanisms: the I-monohalocarbons, the I-polyhalocarbons and the Br-polyhalocarbons. In the Discussion, only the dominant or most important halocarbon of each group is discussed. In this way, the reader does not drown in all data, but is presented with the results that are of interest. This is a clear-cut structure and the reader is presented with valuable information on production and removal processes of these selected compounds.

We are very grateful to the reviewer for their positive overview of the manuscript.

[Referee] i) Tables 2, 4 and 5 present correlations of relationships between halocarbons and selected biological parameters. However, it is not possible to deduce whether the relationship is positive or negative. Moreover, the authors themselves seem to be confused too (see comments below). Please add this information in the tables and make explicit in the text when dealing with a positive and when with a negative correlation.

Tables 2 and 5 have had negative signs added to data that displays an inverse correlation, ensuring the reader can be certain of the nature of the relationship. All data in Table 4 is positively correlated so no alterations have been made. Furthermore, there was indeed some ambiguity in a small portion of the text with regard to the nature of the relationships:

P8216, L7-11 now reads:

"Furthermore, concentrations of CH_2I_2 were strongly, and often significantly, correlated with a number of biological parameters. Shown in Table 5, CH_2I_2 was closely positively correlated with both chl a and total bacteria for the whole experiment, whilst close positive relationships with the phytoplankton pigments fucoxanthin and peridinin were observed during PIII. Taking into account the relationship between CH_2I_2 and biological parameters, the possible reasons for an increase in net production of CH_2I_2 in response to increasing pCO_2 will be explored in the following section".

[Referee] ii) Section 4.1.2: 1) The text on p. 8216 suggests that Table 4 indicates positive correlations only, but figures 3 E-H show a more complex relationship and on p. 8217 the relationship between CH2I2 and bacteria is deemed negative, whereas it clearly is positive. Please clarify. 2) The ratio of CH2I2 to a number of biological parameters were found to be correlated with pCO2. This is however due to the fact that these biological parameters vary with pCO2, not CH2I2! This ratio correlation can be found for any parameter that did not change with pCO2 and does not give causal information on trends in CH2I2. Hence fig 5 plus discussion can be deleted (btw: 5c does not exist).

Figure 5 and accompanying text has been removed.

[Referee] 3) The suggestion that up-regulation of CH2I2 might be an indication of an antioxidant function and hence perturbed cell physiology with increasing pCO2 is interesting, but also puzzling, because, if anything, one would expect a reduction in ROS production with increasing pCO2, not an increase. So, are we looking here at the response to an unknown stress factor inducing ROS? This nuance should be added when discussing this potential function.

We have taken the reviewers comments into consideration and agree that this nuance should be included. With some re-wording and additional text, the sentence now reads:

"Therefore the up-regulation of CH_2I_2 production seen here in response to the altered seawater carbonate chemistry, <u>or indeed some other unidentified stressor</u>, may be indicative of an adaptive response due to perturbed cell physiology amongst the plankton community".

[Referee] iii) Section 4.1.3, last paragraph: This is a relatively lengthy discussion of observed differences in CHBr3 between mesocosms at 1 point in time (t21). The difference is attributed to bacterial activity and not numbers, since those do not deviate. But if bacterial activity is so important from one day to the other, than why doesn't it get any attention in the rest of the discussion? If bacterial activity does change at other instances, without any impact on halocarbons, that would be interesting to know as well and would put the importance it is given now into perspective.

Taking the reviewers comments into consideration, we now feel that this paragraph is unnecessary – and perhaps does not add a great deal of insight in to the discussion. Our intention was to attempt to explain the elevated concentrations of $CHBr_3$ in M1 relative to the other mesocosms, and there is indeed some apparent relationship between the net change in concentration of $CHBr_3$ over certain short periods of the experiment, and bacterial biomass production. However, as this link doesn't hold up at any other instances than those mentioned in the text, the importance is probably low. Thus we have removed the aforementioned paragraph from the body of the text.

[Referee] - Sections 2.5.3 and 2.5.4 are in fact subsections of 2.5.2. Please renumber. Also I was wondering whether this means that the samples for "chl a and additional phytoplankton pigments" were NOT taken from the same cast used for halocarbons. And if not, do you expect any differences. It would make sense to write a paragraph on sampling the ancillary parameters right under 2.5 and then delete 2.5.2.

The reviewer is indeed correct – and it appears the confusion over these sub-sections arose post-submission. Changes have been made to the text to ensure clarity, and are presented as follows:

2.5 Ancillary measurements

Chl a and additional phytoplankton pigments

Microbial abundance

Total bacterial abundance

All biological measurements were taken from the same cast as for halocarbons, and this has been clarified in the text, with a sentence added under section 2.5.

[Referee] - In a printed version, figure 2 is far too small. Please reformat.

In the format of a discussions paper, this figure is indeed too small to clearly see the detail. However, for full publication the figure would work if it was presented in landscape format. Equally, it would also work in portrait format so we have produced two versions of the figure, and we will discuss the best solution with the editorial support team.

[Referee] - Section 3.2: "To simplify analyses and to give an overview of general trends, the halocarbons were assigned to three groups. . ." Not only that: you also present means of all treatments. Please add this in the wording, since it is now stated nowhere.

The reviewer correctly points out that it is not clear in the body of the text that the means of all mesocosms are used. Text now reads:

"To simplify these analyses and to give an overview of general trends, halocarbons <u>concentrations were averaged across all mesocosms and</u> assigned to three groups based on their common biological production pathways (Manley 2002):

[Referee] - Section 3.3: What is exactly meant with "cumulative concentration"? How was it calculated? And why is it a better representation than concentration only? R is exactly the same.

Cumulative concentrations were shown to give an indication of net production over the course of the experiment. However, as net production is also shown in the figure and the relationship with pCO_2 is indeed the same as for mean concentrations, cumulative concentrations have been removed from this figure. Cumulative flux has also been removed, so now only mean flux is shown.

[Referee] - Discussion, 3rd line: Table 6 is in fact the 2nd Table referred to. Please change in order of appearance in the text.

We thank the reviewer for noticing this discrepancy in table numbering. Table 6 is now presented as Table 2, and all other table numbers have been altered accordingly.

[Referee] - Page 8215, Line 23: There are several reasons why UVR is relatively low in June at Ny-Alesund, but not the solar zenith angle, which is at its highest at that time of the year. Delete that part of the sentence.

We acknowledge that the solar zenith angle is at its highest in June – however, due to the high latitude of Ny-Alesund, the angle is still relatively low and this would result in little solar UV entering the mesocosms from above. However, we are happy to delete the suggested part of the sentence to avoid confusion from the reader.

[Referee] - Figure 6A: Y axis indicates "net loss rate minus flux" but figure legend indicates net loss rates only. Please clarify/change.

"minus flux" has been removed from the axis title.

[Referee] - The very last sentence of the conclusions suggests an increase in importance of halocarbons with retreating sea ice, but there is really no evidence for that presented in this paper. Firstly because there is no indication of increased production of halocarbons with increased biological productivity, which is predicted to take place with the loss of sea ice. And secondly because this paper doesn't show what the effect of sea ice on halocarbon production is. This work enhances our understanding of halocarbons all right, but not in the context of reducing sea ice.

We agree with the reviewer that this paper does not show the effect of changes in biological productivity on halocarbon production. We have removed the reference to changes to biological activity, and it now reads as follows:

"The role of halocarbons in Arctic atmospheric chemistry may increase in importance in the coming decades due to increases in open water with the loss of sea ice (Mahajan et al. 2010, Stroeve et al. 2011); this work enhances our understanding of the marine production and cycling of halocarbons in a region set to experience rapid environmental change".