Reply to Referee #1

We thank referee#1 for his/her comments. Below you find the detailed answers (normal font) to the issues raised by the reviewer (`typewriter`). Added text blocks for the revised version of the manuscript are written in italics, citations of the original manuscript are enclosed in brackets [].

`This paper describes a study in which the authors test the sensitivity of simulated CH3I to different production mechanisms in seawater. In general, I find this study to be marginally valuable, as there are enormous sources of uncertainty and unaccounted for variability that are not addressed. As such, it is not quite possible to sustain the conclusions in this paper based on the interpretation as presented, statistical or otherwise.`

`Major corrections are necessary to permit publication.`
`General Comment:`
`1) The text is in many ways poorly written. It could be that the authors are non-native English speakers. In any case, many parts of the paper need extensive revision for clarity, correct grammar and punctuation. This undermines a good deal of my ability to properly interpret the material. Unfortunately, I am not patient enough to provide point-by-point recommendations for correction. In instances where the errors are egregious I have included specific comments.`

We will improve the English language in the revised version of manuscript.

`2) This paper does, in my view, present a solid review and evaluation of available observations. This is a valuable part of this study.`

`3) It may be my fault, but I cannot find where the atmospheric conditions are described. What I gather is that time-invariant atmospheric concentration were used. Why? The atmospheric boundary conditions must have an enormous impact on the MBL simulation results; and given the reaction rates in the atmosphere, and the overall lack of observations, some effort to evaluate the impact of the use of fixed conditions must be included.`

As noted on P6L24 (P17555L24) & in sect 2.3 we use the global climatological field of observed atmospheric CH3I concentrations compiled by Ziska et al. (2013), specifically we use their results obtained by the robust fit method interpolated onto the model grid. In most oceanic region the ocean is strongly over-saturated with CH3I and atmospheric concentrations will be hence dominated by emissions (i.e. marine production). Of course in regions where the ocean gets under-saturated, the usage of a time-invariant field of observations is inappropriate. This occurs in the North Atlantic in boreal winter. As we tried

to clarify on P24L28 (P17573L28) P25L1-3 (P17574L1-3) atmospheric concentration data are insufficient to deduce the temporal variability there. Therefore we like to postpone any detailed analysis to a later state, when a fully coupled model system is ready to use, which is in progress and not available yet.
As discussed on P25 (P17574) we believe that the feature of reversal is robust, as the atmospheric concentration value we use here, are lower than previously measured concentrations (due to the spatial interpolation).

We will add a short explanation on P6L24 (P17555L24) in the revised version of the manuscript:
[.. using a time invariant field of atmospheric concentrations] *(Ziska et al. (2013), robust fit interpolation method,* [see Section 2.3] *on data basis).*

4) Would it not be relevant to include atmospheric observations to confirm local air-sea exchange estimations?

Our boundary conditions are based on globally extrapolated observations. Details can be found in Ziska et al. (2013).

5) How does the uncertainty compare between the source functions, their dependencies, air-sea exchange limitations, the resulting CH3I concentrations, and their deviations compared to observations? What is the potential impact of atmospheric oxidation?

Atmospheric oxidation of methyl iodide implies that the substance will not accumulate in air (life time of 4-5 days) and hence concentrations of CH3I in the marine boundary layer are largely determined by emissions; temporal variability can be expected to be similar in atmosphere and ocean. Indeed the ocean is strongly over-saturated in most regions. Any uncertainty related to atmospheric oxidation rates is thus not relevant for marine CH3I concentrations and emissions.
We will include an analysis of the sensitivity of the choice of input parameters and forcing (see below, reply to comment 11).

6) Are vertical gradients considered in the analysis? With soluble species such as CH3I, local vertical gradients can be large, no?

We are aware that the CH3I vertical distribution is spatially and temporally variable (as we have discussed in our analysis of methyl iodide concentrations in the tropical eastern North Atlantic in Stemmler et al. (2013)). However, the analysis of vertical distribution patterns of CH3I is out of the scope of this manuscript. Here, we want to focus on surface concentrations resulting from different source processes and subsequent emissions.

7) Is mixed layer depth not a factor here? It would be a means of reconciling most of the dependencies controlling the production variability. Mixed layer

depth, combined with the vertical structure of production and concentration fields would be a useful analysis.

We are not really sure whether we got the point. Generally, the mixed layer dynamics will of course also influence the dynamics of CH3I. In the case where CH3I production takes place below the mixed layer (as in the subtropical oligotrophic ocean for the biological pathway) a strongly stratified ocean would "shield" methyl iodide from gas exchange. A deeper and stronger mixing would bring the subsurface CH3I to the surface. As a result gas exchange and accelerated degradation due to higher temperature will take place. Thus a deepening of the mixed layer leads to a reduction of the residence time and a shoaling to potentially longer residence times (in regions with a subsurface CH3I maximum). In turn when CH3I is produced within the mixed layer, a deepening of the mixed layer would lead to dilution, i.e. to reduced concentrations, reduced outgassing, and thus a longer residence time. We will not include a deep analysis of the impact of the mixed layer dynamics, but where relevant we will refer to these effects (P9L21 (P17558L21), see specific comments below).

8) Error/Uncertainty estimates of model results are not included. For a model study like this, it is essential. As stated before, this is a major short-coming of this paper.

We add model experiments to address this issue. See below (reply to comment 11).

9) There is excessive reliance on supplemental information to support fundamental assertions. For example, P11L6-7 (P17560L6-7) (see specific comments, below).

We can not include the whole set of cruise data shown in the SI into the main text. To provide more information in the main text we follow the suggestion of the reviewer (in comment 10) and include more aggregated figures on model evaluation (see below) and refer to the figure on P11 L6-7.

10) Some specific non-spatial comparisons of model to obs. data should be added:

For example, scatter plots would be of enormous benefit to interpretation of mapped & tabulated results. It is not possible to discern the models performance from the difficult-to-interpret maps of cruise tracks.

We agree with the reviewer, that an aggregated figure with a comparison between model and observations is missing. However, we have to refrain from showing a scatter plot, as it would not be adequate, since the observations are not available in a gridded format like the model results. Due to the model's relatively coarse horizontal resolution very

often several observational data points fall into one grid cell. For a scatter plot either these point data would have to be averaged or arbitrary data points need to be excluded from the analysis. Instead show Box-Whisker plots for the Atlantic Ocean (see below), that aggregates data into 10 degree latitudinal boxes and allows for an easy comparison of all experiments with observations over a large region in one figure. For these plots all observations within a 10 degree latitudinal zone (or mean model results within 1 degree of the respective locations) are plotted as boxes with size characteristics obtained from the statistics of the data (model results). The width of each box is determined by the 25 % and 75 % percentiles, furthermore it shows the median of the data, and outliers, i.e. any data smaller or larger than 1.5 times the difference of the 75 % and 25 % percentile.

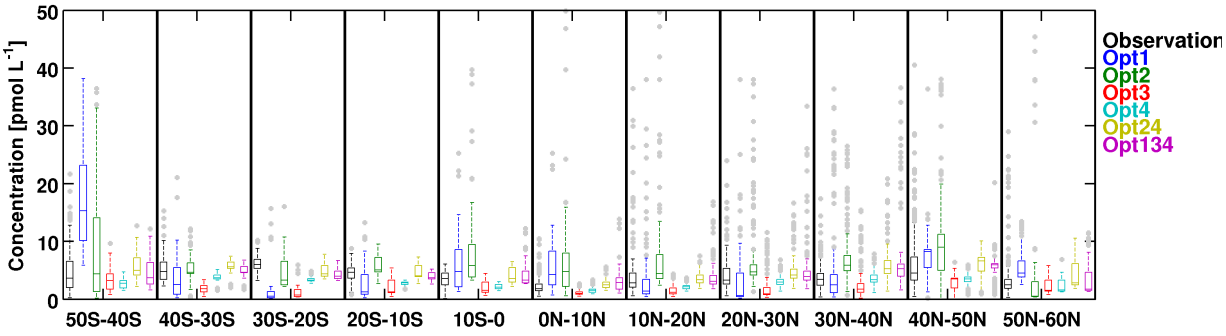

Figure 1: Box-Whisker plot of simulated and observed surface ocean CH3I concentrations [pmol L-1]. Box widths are determined by the 25% and 75% percentile of data within each 10 degree latitude box, outliers (gray) are located outside 1.5 times the differences of the percentiles, the middle line of each box shows the median. Simulated concentrations are averaged over 1 degree boxes around the location of observations.

In the revised version of the manuscript on p12l5ff this figure (there Fig.2) will be referred to:
[The simulated surface methyl iodide concentrations are compared with observations (Figs.1, *2*, 8, and S1-S25). Generally the model represents methyl iodide observations well (*Fig. 2* and Supplemental Information Figs. S1-S25).]

11) The closest observed value method outlined starting at P13/L26 is interesting but,in my opinion, flawed. For this analysis to have merit, an uncertainty analysis of all of the corollary data, dependent and independent sources of variability, compared to observations would have to be included that is able to state that the difference between model & observation is (1) statistically significant, and (2) to what degree the results are driven by difference in model skill in reproducing the CH3I production drivers (e.g. DOC, phytoplankton, dynamics, gas-exchange, etc.). The authors start to perform this analysis in

Section 3.2, but the interpretation is weak and is not used in the interpretation of the results from the closest observed value analysis.

We agree with the reviewer that information on uncertainty is missing but needed and that the discussion of the impact of driving parameters (e.g. DOC, PP) is fragmentary to explain the differences between simulated and observed concentrations.
One of the fundamental assumptions of the model is that for any CH3I source process only one (set of) production rate (ratio (s)) is used globally. This implies that the input parameter setup we derived for the Northeastern tropical Atlantic (Stemmler et al. 2013) can be tested on global scale (following the logic of the model). In Stemmler et al. 2013 we derived the input parameters from a numerical parameter optimization, hence the chosen set of input parameters represents the 'optimum' parameters to reproduce observed CH3I concentrations.
Therefore we can not easily come up with an underlying probability density function (PDF) for all CH3I production rates to perform a Bayesian uncertainty analysis (based on Monte-Carlo-simulations of the input parameter used in several model runs); we cannot easily derive quantitative information on the significance of differences between simulated and observed concentrations. This is even more complicated for biogeochemical impact variables such as DOC or phytoplankton, as these can not be modified independently. To cope with this issue and to provide information on the sensitivity of simulated methyl iodide surface concentrations to the choice of production rates and (a)biotic variables we performed a set of additional experiments.
For each experiment that includes only one source process depending on a single production rate (ratio) (Opt1,Opt3,Opt4), we performed two additional experiments with the production rate (de-)increased by 10% (called Opt1,3,4m and Opt1,3,4p, where 'm' and 'p' stand for 'plus' and 'minus').
Another fundamental assumption of the model is, that large scale features in the CH3I concentrations are unaffected by the interannual variability of environmental conditions, which allowed us to use the climatological OMIP (Ocean Model Intercomparison Project) forcing. To address the impact of different environmental conditions, we performed a model experiment in which the model is forced with NCEP reanalysis data in a transient experiment for the years 1959-1973 in contrast to the OMIP climatological forcing used in the previous experiments. Furthermore, this forcing is temporally higher resolved, as it includes a diurnal cycle based on 6h-ly values, in contrast to the daily mean data used in OMIP. Also here the 'standard' setup of rates used in HAMOCC (plankton grazing, mortality, growth) is used instead of the one presented in Stemmler et al. 2013. This introduces modifications of the relevant biogeochemical (e.g. phy, doc) and abiotic (radiation, temperature, wind speed) variables. Large scale features that are determined by characteristics of the physical model (circulation patterns and resulting limitations (e.g. nutrient trapping in the tropical Pacific) are unchanged as the identical model (MPIOM) was used to derive tracer advection and diffusion. This experiment includes only production of methyl iodide from photochemistry (Opt4), i.e. production depends only on incoming ra-

diation, sinks (degradation and gas-exchange) depend on temperature and wind speed. We did not include biological production (Opt1) and photochemical production from SLDOC (Opt3) in the experiment, because analysis of the previous experiments already revealed that both of them show unrealistic gradients (from oligotrophic subtropical gyres towards more productive ocean regions). These features are inherent also in an NCEP simulation, as they are shaped by the large scale oceanic circulation (independent from forcing).

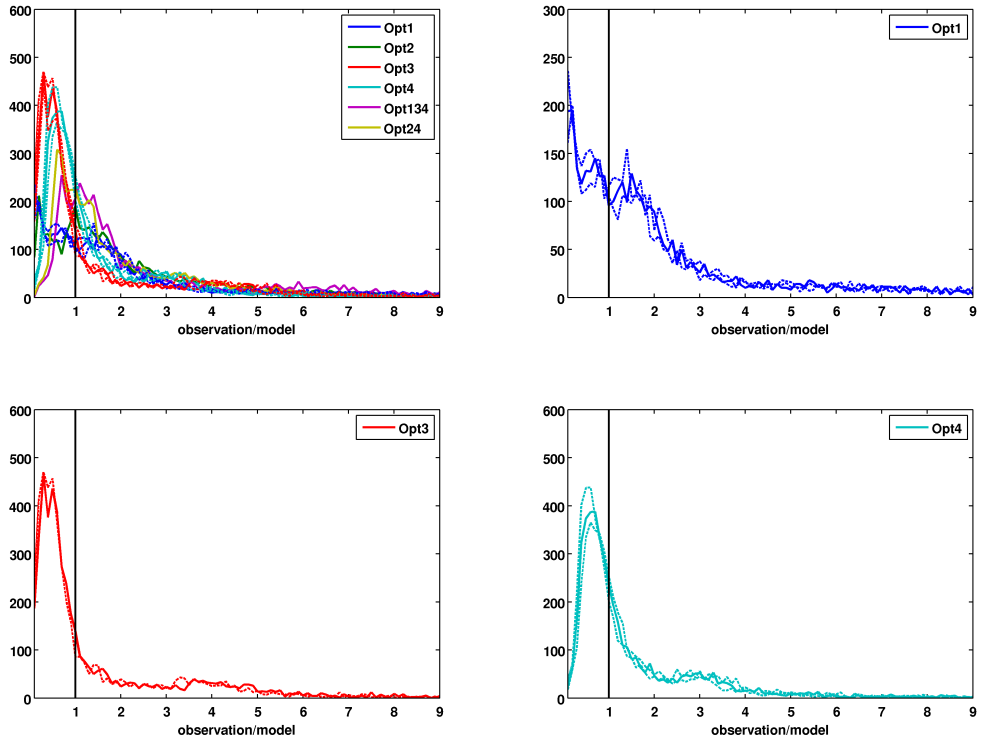

Figure 2: Histogram of the ratio between globally observed and simulated surface ocean methyl iodide concentrations (R=obs/model) for all original experiments and Opt{1,3,4}{p,m} (upper left), Opt1 and Opt1p & Opt1m (upper right), Opt3 & Opt3p & Opt3m (lower left), and Opt4 & Opt4p & Opt4m (lower right).

The spatial distribution is unaffected by the choice of input parameters (Fig 3&4), and differences among the experiments are larger than differences in the uncertainty experiments, i.e. when production rates are varied by 10 %(the p and m experiments (see Fig.3, 4)). Therefore, the shape of the PDF of the ratio observation/model is the same (Fig 2), which means the relative performance of the experiments is largely unaffected by the input

parameters, as long as deviations from the optimum value are small (i.e. when production rates are e.g. 1000 times higher the performance will be lost even for the most successful experiment). Thus, the identification of Opt4 as the most successful experiment is robust and not by chance, though we can not give the exact measure of statistical significance, here. By using transient NCEP 6-ly instead of a climatological daily mean OMIP forcing, and looking into individual years, we can asses the robustness of the model results towards driving variables (here radiation). Similar to the results of the p & m experiments the spatial distribution and the PDF of the ratio observation/model do not change considerably (Fig.s 5,6). This again means the identification of Opt4 as the most successful experiment is robust. The reason for this is that the large scale features in the concentration distribution are more influenced by the choice of the source process rather than by the choice of the parameter values. More specifically, the experiment that are directly or indirectly driven by total primary production (Opt1, Opt3) are less successful because of the deviation arising from the spatial distribution patterns of primary production (low in subtropical gyres) and not from production ratios.

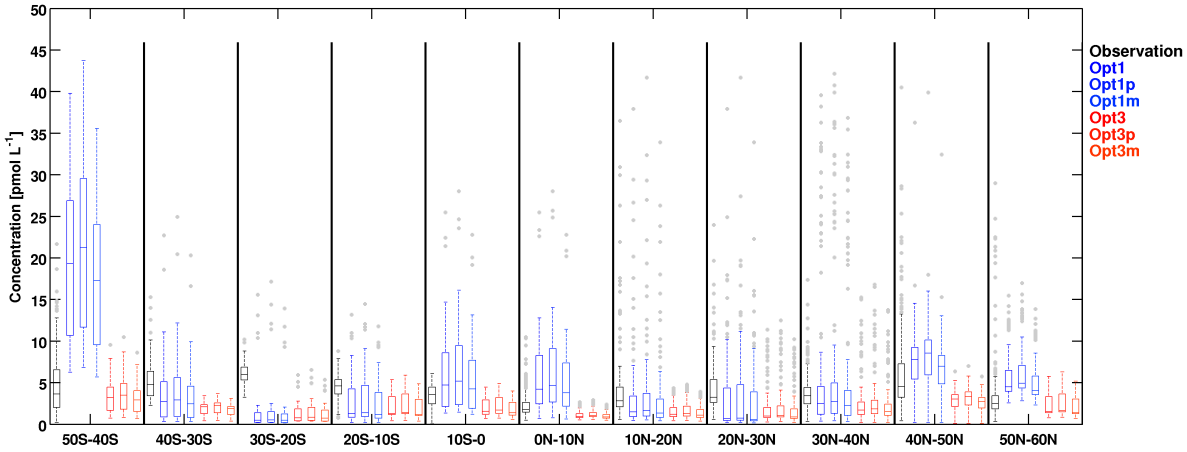

Figure 3: Box-Whisker plot of simulated and observed surface ocean CH3I concentrations [pmol L-1]. Box widths are determined by the 25% and 75% percentile of data within each 10 degree latitude box, outliers (gray) are located outside 1.5 times the differences of the percentiles, the middle line of each box shows the median. Simulated concentrations are averaged over 1 degree boxes around the location of observations.

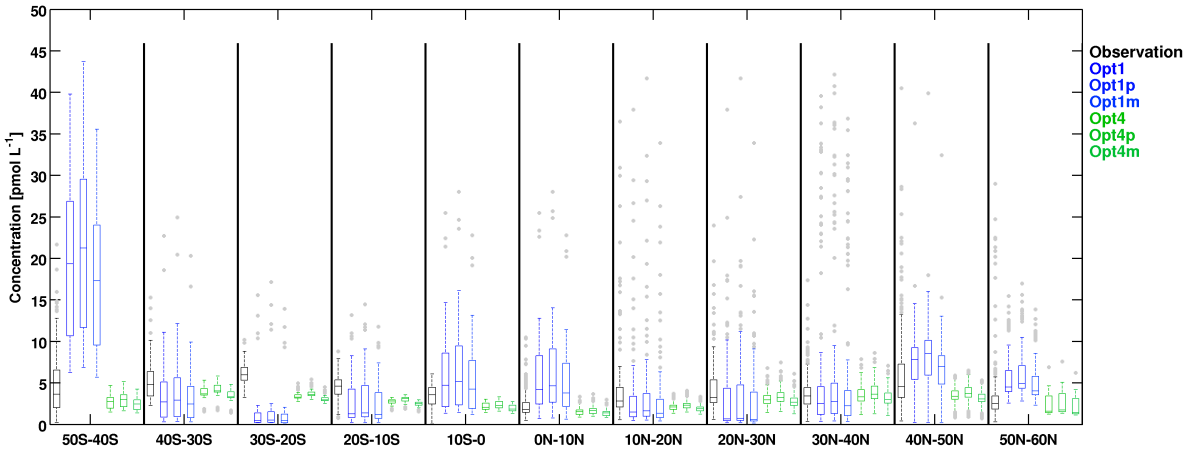

Figure 4: Box-Whisker plot of simulated and observed surface ocean CH3I concentrations [pmol L-1]. Box widths are determined by the 25% and 75% percentile of data within each 10 degree latitude box, outliers (gray) are located outside 1.5 times the differences of the percentiles, the middle line of each box shows the median. Simulated concentrations are averaged over 1 degree boxes around the location of observations.

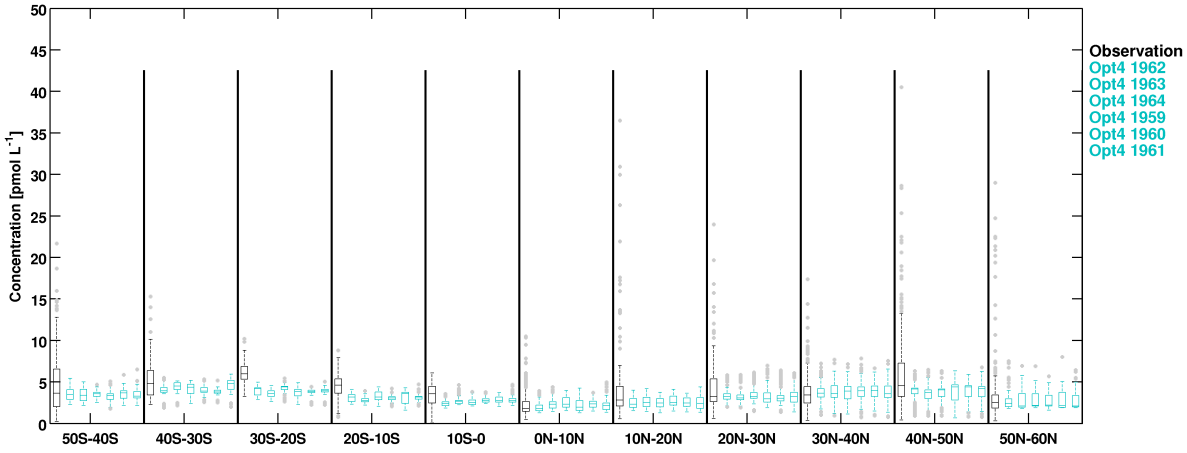

Figure 5: Box-Whisker plot of simulated and observed surface ocean CH3I concentrations [pmol L-1]. Box widths are determined by the 25% and 75% percentile of data within each 10 degree latitude box, outliers (gray) are located outside 1.5 times the differences of the percentiles, the middle line of each box shows the median. Simulated concentrations are averaged over 1 degree boxes around the location of observations.

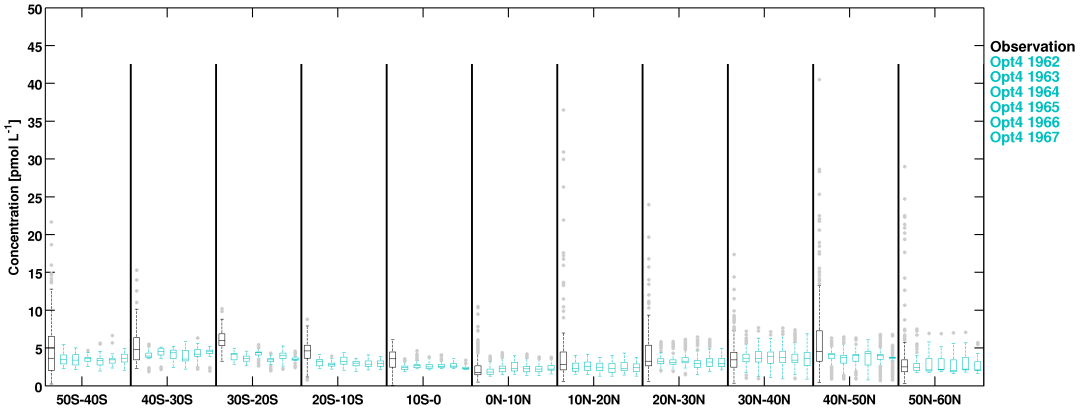

Figure 6: Box-Whisker plot of simulated and observed surface ocean CH3I concentrations [pmol L-1]. Box widths are determined by the 25% and 75% percentile of data within each 10 degree latitude box, outliers (gray) are located outside 1.5 times the differences of the percentiles, the middle line of each box shows the median. Simulated concentrations are averaged over 1 degree boxes around the location of observations.

We will add the following in the revised manuscript P16L11 (P17565L11):
[Opt3 furthermore underestimates observations more strongly than Opt4 and Opt2, indicated by the highest median ratio of observations and simulated concentration.] *A variation of the production ratios by 10% in Opt1, Opt3, Opt4 leads to the same spatial distribution (Fig 3,4) and the PDF (probability density function) of the ratio observation/model (Fig 2) is unchanged. This is also true when a different forcing (transient NCEP 6h data instead of climatological daily mean OMIP) is used in Opt4 (e.g Fig. 5). Thus, differences in the RMSD and the ranking (Tab 3) of the experiments are robust and driven by the spatial distribution of the concentrations rather than by the choice of production rates. In particular, Opt1 and Opt3 are least successful, because their spatial distribution is determined by primary production, which shows features that are not present in global patterns of observed methyl iodide. In contrast to model results, observations do not show low concentrations in oligotropic subtropical gyres and high concentrations in productive regions. However, one has to keep in mind, that halocarbon production rates are different for different phytoplankton species (Moore and Tokarczyk 1993, Hughes et al 2006, Smythe-Wright et al. 2006, Brownell et al 2010), hence community composition differences would lead to changes in the bulk CH3I production rate and distribution patterns. The inhomogeneity of the community composition and CH3I production rate implies that also chlorophyll-a may not be a good predictor of CH3I production (see also Section 3.2). The success of Opt4 over Opt3 shows that the representation of DOC indeed strongly influences the ability of the model to reproduce observed concentrations when considering photochemical production*

alone. Where Opt3 shows minima due to low DOC, Opt4 shows maxima due to high insolation. These differences are higher than differences that arise from variable insolation (i.e Opt4 with OMIP forcing compared to Opt4 with diurnal NCEP forcing in different years). In reality, most likely biological and photochemical production occur at the same time. In [the next step we consider the hypothesis ..] (P16L12, i.e. P17565L12)

Specific comments:
P8/L11: Define nutrient levels. What is plenty?

Will be rephrased:
[Two experiments were performed, one with normal biological production, in which the ratio between CH3I production and primary production is kept constant (Opt1), the other with a varying ratio (Opt2), which is high in oligotrophic oceans and low] *where phytoplankton growth is hardly nutrient-limited.*

P8/L25 - P9/L2: This should be in methods.

It needs to be kept here, since this is a model result: kpp varies as a function of the nutrient limitation factor of phytoplankton growth, which in turn depends on iron, phosphate and nitrate, which are prognostic tracers of the model.

P9/L9: Why is production reduced in the storm tracks?

The reviewer refers to:
[As production is reduced in the windy storm track regions of the Southern Hemisphere and intensified in the warmer subtropical regions the relative importance of outgassing is reduced compared to Opt1 (in the global budget, see Table 2), whereas the temperature- and light-dependent degradation processes gain in importance.]

Due to stronger mixing which favors nutrient supply from deeper ocean layers to the surface, in the SH storm tracks phytoplankton growth is not strongly nutrient-limited. This implies that kpp, the CH3I production ratio, is lower in Opt2 and less CH3I is produced compared to Opt1 (which uses a constant kpp). In turn kpp is higher in the oligotropic ocean due to nutrient limited phytoplankton growth, and hence CH3I production is higher than in Opt1. This means a relative shift of production from cold and windy to warm and bright regions and [the relative importance of outgassing is reduced compared to Opt1 (in the global budget, see Table 2), whereas the temperature- and light-dependent degradation processes gain in importance.]

To make this more clear in the revised version we will add:
[As production is reduced in the windy storm track regions of the Southern Hemisphere] *(due to the lower CH3I production ratio in this biologically productive region)* [and intensified in the warmer subtropical regions the relative importance of outgassing is reduced

compared to Opt1 (in the global budget, see Table 2), whereas the temperature- and light-dependent degradation processes gain in importance.]

P9/L16-17: By definition, isnt Labile & semi-Labile DOC bioavailable? Is microbial uptake not considered in the cycling?

We are aware that (semi-) labile DOC is by definition bio-available, as microbial uptake leads to the short residence times of DOC in the ocean. This is also represented in the model. We included this redundant clarification to remind the reader of the different experiments, which are only briefly explained in this manuscript, but detailed in Stemmler et al. 2013.

P9/L27: It seems youre stating that CH3I produced from refractory DOC? How is this possible? Is it a recognized mechanism?

As stated in the method section, produced from 'refractory DOC' implies production from the photochemical production with unlimited supply of DOC. We chose an experimental setup that includes the semi-labile DOC from HAMOCC and a constant DOC pool for the following reasons:

According to Moore & Zafirou 1994 photochemical production of CH3I most likely occurs via radical recombination of CH3 and I, whereby the methyl radicals may origin from photolysis of humic material. Incubation experiments with filtered seawater show production of methyl iodide when the samples were irradiated (e.g. Happel & Wallace 1996, Richter & Wallace 2004). However, speciation of dissolved organic carbon from these experiments is not available. From this information, it seems reasonable to build a parametrization of the photochemical production pathway on radiation and dissolved organic carbon (DOC). In nature, there are numerous bioavailable and refractory dissolved organic carbon species with different characteristics. HAMOCC only considers one DOC pool that has a life time of a couple of months. Thus, we decided to set up one additional experiment that does not use HAMOCCs description of DOC. For simplicity and to be consistent with Stemmler et al. 2013, we decided to introduce a constant DOC pool, rather than implementing a new DOC tracer with a longer lifetime. We call it refractory DOC, as extremely long life times would most likely result in an almost uniform distribution of DOC. In fact, the experiment just reflects an unlimited source of DOC.

P10/L21: In my view, mixed-layer depth is a prime candidate for these results.

We agree with the reviewer that the mixed layer may play a role in prolonging the residence time of CH3I in the Arctic and in affecting the timing of maxima. In the Arctic CH3I production in Opt1 is largest within the mixed layer. A deepening of the mixed layer would lead to dilution, i.e. reduced concentrations and reduced outgassing, thus a

longer residence time. We will add this in the revised version of the manuscript:

[The reasons for this are small losses, i.e. reduced outgassing in summer when the wind speed is low and slow chemical reactions (hydrolysis, nucleophylic substitution) at cold temperatures.] *Furthermore, methyl iodide production occurs within the mixed layer. Deepening of the mixed layer in windy seasons lead to dilution of concentrations, which is not compensated by production, and thus results in a longer residence time.*

P10/L22: Define significant. Is there a p-value for this? Also on P15/L6.

As significant is often understood in the statistical sense by readers, which is not meant here, we will replace it in a revised version by considerable.

P11/L6-7: The statement that the model reproduces observations well is a qualifying statement that should be avoided unless considerable quantitative interpretation is included. Supplemental information is cited here. But, Id argue this is a fundamental piece of information that should be in the main text.

In our opinion a qualitative statement is important here, because when we discuss the results we need to judge the quality of the model. Not only quantitative measures help, but also some information is needed whether a deviation factor of 3 or 2 is regarded as a good representation; this is what we do by stating matches well.

P11/L26: Dont abbreviate language in main text. (approx. should be approximately). Approximate also suggests a statistic that is not given.

We disagree on this. Approximately does not necessarily implies a statistical meaning, but can simply mean 'not exactly'. We will use the long version in the revised manuscript.

P12/L1: Statement not clear.

The reviewer refers to:
[It is only at the Equator where this experiment (Opt2) still overestimates observed concentrations by a moderate factor of approximately 1.3]

It will be rephrased:
However, similarly to Opt1, this experiment (Opt2) overestimates observed concentrations at the Equator [by a moderate factor of approximately 1.3.]

P13/L20-24: The authors assert that seasonality produced by the model but not seen in the observation is the fault of the data. It is reasonable to cite the need for more observations; but if a model result isnt supported by the obs., then it can be argued more likely the result of the model mechanics rather

than lack of observation.

The reviewer refers to:
[For the Atlantic some information on the seasonal cycle of CH3I concentrations is available from Wang et al.(2009). They find maxima in summer in the North East Atlantic south of Greenland (compare Figs. S21-S23). The model experiments Opt1 and Opt2 show maxima in spring, only the experiments that are dominated by photochemical production show maxima in summer, with a similar magnitude in and difference between the seasons as in the observations.]

Here we list first what observations tell us about the seasonal cycle and what different model experiments show. We do not include any conclusion here, but only state that it is only the photochemical experiments that are able to reproduce the observed seasonality. We do not indicate any fault in the data. In contrast, the obvious conclusion from this statement is, that the experiments with photochemical methyl iodide production are more realistic than with biological production if temporal variability is considered.

P13/L23-24: This statement is speculative, and not supported directly by results or analysis.

The reviewer refers to:
[The model experiments Opt1 and Opt2 show maxima in spring, only the experiments that are dominated by photochemical production show maxima in summer, with a similar magnitude in and difference between the seasons as in the observations.]

This is no speculation, but the description of a model result. We could support this by showing it in a figure (e.g. the one below) or table, but we decided to only describe the result.

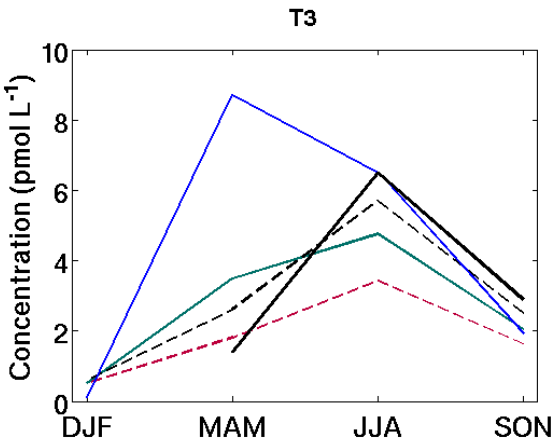

Figure 7: Simulated (red dashed line Opt134, black dashed line Opt4, green line Opt24, blue line Opt1) and observed (black solid line) CH3I concentrations in the Northern West Atlantic (location T3 in Wang et al 2009) [pmol L-1].

P15/L7-12: This is good. Tuning is a serious and unaccountable problem in modeling.

Section 3.2: The results from this section should be tabulated. Perhaps an ANOVA would help.

All results of this section are depicted in Figure 10. An alternative way of illustrating them would be listing all numbers in a table, but considering that we calculated 3 kinds of correlations (with phy, sst, rad) for 6 experiments and 12 months (upper part) and 3x12 additional correlations, a table would make it hard to digest the information. All of the correlations shown are significant ($p<0.05$).

P16/L5: Can radiation and phytoplankton be considered independent variables? If not, the a correlation analysis is invalid.

Radiation and phytoplankton are dependent variables. In fact, none of these variables are truly statistically independent: temperature depends on radiation and phytoplankton depends on both radiation and temperature, also methyl iodide depends on all of these parameters. But, the goal of this section is neither to generally assess the covariability of abiotic and biotic variables and methyl iodide nor to conclude about causal relations from it, but to illustrate the weakness of this widely used method. This way often methyl iodide sources are identified from in-situ measurements of concentrations and other variables. Our aim in contrast is to inform about the weakness of this method. We agree with the reviewer, that it needs to be additionally mentioned that the prerequisites for applying

this method (independence of the variables) are also not given. This will be done in the revised version of the manuscript.

Table 3: The table is hard to interpret. What is being shown in the 1st three rows? What are the units? I understand that the caption states this, but it should be clear in the table. Also, is a median RMSD the median of the root mean squared deviation? That seems self-contradictory. If the data are not normally distributed, is a RMSE even valid? I may be wrong here. Apologies if I am; but perhaps this should be clarified in the text.

We will modify the wording in the table:

Table 1: Global fraction of observations best presented by the respective model experiment (as shown in Fig.9 for individual locations and seasons), considering only single source experiments, only mixed sources experiments, and all experiments (upper part). Global root-mean-square deviation (RMSD, $[pmol\ L^{-1}]$), root-median-square deviation (RMSD(median), $[pmol\ L^{-1}]$), and global median of the ratio $\frac{observation}{model}$ (lower part).

Source	biological		photochemical		mixed	
ID	Opt1	Opt2	Opt3	Opt4	Opt134	Opt24
Percentage of simulated data closest to observations:						
Including only single source exp.	13.12%	24.15%	16.46%	46.27%		
Including only mixed source exp.					39.31%	60.69%
Including all exp.	7.63%	16.56%	12.14%	34.27%	18.17%	11.23%
Characteristics of the comparison of model and observations:						
Global RMSD $[pmol\ L^{-1}]$	7.78	10.11	2.97	2.63	3.14	3.21
Global RMSD (median)$[pmol\ L^{-1}]$	1.04	0.65	0.50	0.35	0.60	0.52
Median ratio $\frac{observation}{model}$	2.14	1.74	3.42	2.01	1.24	1.49

Showing both the median and the mean of the root deviations is done to characterize the pdf probability density function (see above Fig.2); differences imply a non symmetric distribution of the deviations (see additional information above (reply to comment 11)). We will mention this in the revised version of the manuscript.

References

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