#### Anonymous Referee #1

This manuscript addresses the oceanic sources of three volatile halocarbon compounds that contribute to potentially important aspects of atmospheric chemistry. The paper provides information on the distribution and air-sea flux of these compounds with a focus on the Mauritanian Upwelling region as a source of high amounts of these compounds. Measurements of halocarbons in seawater are relatively rare and even less common are reports that combine these with atmospheric measurements and the environmental factors, including biological variables that may influence their production and emission. In this respect the manuscript is appropriate subject matter for ACP and is a potentially valuable addition to the special issue. The manuscript aims to compare distributions of the halocarbons between oceanic waters and the more productive, coastal upwelling regions and provides estimates of the air-sea fluxes in each location. In general the authors do a good job of this aspect of the manuscript. What is less convincing is the determination of the sources of the compounds which relies almost entirely on correlative analyses between, in some cases, poorly constrained proxies of biological variables. I think this evidence is over-interpreted with little explanation of the uncertainties involved or potential alternative sources not tested. The manuscript is well written and results are well presented. An important point is the extent of overlap, with the Fuhlbrügge et al. submission to the same Special Issue; particularly in relation to the conclusion regarding the MABL height and its impact on atmospheric mixing ratios.

We thank referee #1 for the very helpful input and suggestions. We agree that the biological variables we used cannot completely resolve the sources and sinks of the measured halocarbons although we believe that our approach shows a general relationship between bromocarbons and biology. Nonetheless, we weaken the relevant passages, and emphasize the limitations of our methods, especially with respect to the reviewer's specific concerns and comments.

To avoid the potential overlap between the two companion papers we carefully check our manuscript again to address the reviewers concern. Just for clarification, while Fuhlbrügge et al. (2013) is concentrating on the meteorological constraints on the distribution on atmospheric halocarbons, our manuscript focuses on the sea surface concentrations, the emissions and their possible contributions to the atmospheric mixing ratios. We think that

both manuscripts complement each other and together draw a picture of the complex interactions in the investigation region.

#### Specific concerns:

## 1. P19706 2.1 L19. Was an internal standard used in the analyses and if not, how was drift in instrument sensitivity monitored?

We did not use an internal standard. Our experience shows that the drift of the experimental set up used during DRIVE is low during the four weeks of a campaign. To monitor this drift, one of the dilutions we prepared for the calibrations which is in the range of the mean concentrations measured was injected in triplicate once a day. Some more information on the calibration procedure is included in the manuscript.

## 2. P19706 2.1 L19. It would be useful to report the purge efficiencies for each of the compounds

We purge 80 mL of sea water 60 min with a stream of Helium of 30 mL min<sup>-1</sup> at 70 °C. We have calculated that the theoretical purge efficiency of  $CH_3I$ ,  $CHBr_3$ , and  $CH_2Br_2$  is larger than 98 % using these specifications. Laboratory and campaign related experience shows that this is matching the observed purge efficiency of our instrumental set up. We add the purge efficiency to the method section 2.1: "80 mL of water were purged with a stream of helium at 30 mL min<sup>-1</sup> at 70 °C in a glass chamber with a purge efficiency of more than 98 % for all three halocarbons."

3. P19706 2.1 L21. More detail is required regarding how precision was calculated from duplicates? Normally, assessment of precision requires at a minimum, triplicate samples. Were true replicate samples analysed – i.e. replicate water samples collected from the moon pool, or were the duplicates from the same sample?

We have determined the precision from duplicates from the same water sample due to time issues. As stated in the paper, we use a purge time of 60 min which unfortunately constrains the number of samples we can analyze per day and is limiting the spatial resolution of

measurements. From laboratory experience, we know that the precision from triplicates from the same sample is in a similar range as the reported precision from duplicates. However, we agree with the reviewer that triplicates would be more suitable. In subsequent campaigns, we have compared two different systems, the GC-MS as described in this manuscript and a coupled GC-ECD with a purge and trap system, measuring the same and parallel samples from the same water supply campaigns for quality control.

4. P19707 2.1 More details of how the intercalibration was performed would be useful, as would evidence of the stability of the individual compounds in the gas samples in the canisters over the approximately 1 month of storage.

- a) Calibration for the air samples was based on comparisons of whole air samples collected and analyzed during the HIPPO airborne research campaign. Replicate samples were collected during the campaigns and analyzed independently in the RSMAS and NOAA laboratories. Calibration of the samples was based on the NOAA standard scale. To ensure the compatibility of sea water samples with air samples, we measured gaseous standard during the cruise with the instrumental set up that was used to measure liquid samples.
- b) Stability and integrity of sample composition during storage is an important consideration in whole air sampling. Often, the stability will depend on canister surface properties, the type of sample (wet/dry), and potentially unknown factors. However, it is not often practical to do stability tests under all conditions of sampling and storage. Thus, after initial testing of canisters, we rely on the measurements themselves to reveal something about the canister stability. We look for consistency between samples collected along a track to reflect sample stability. Our experience indicates that outliers due to compound loss in canisters are usually easy to identify. Other tests we have done are to compare samples collected in the same locations but analyzed by separate laboratories. Typically, sample comparisons between labs show good correlation, with most differences attributed to calibration offsets. One example of a comparison between different canister samplings has been conducted during the TransBrom SONNE cruise showing very good agreement (Brinckmann et al 2012). We have compared some of the compounds directly from the canister measurements (with storage time) to in-situ measurements with the µ-dirac on a recent cruise (Quack and Krüger, 2013) and despite a large standard deviation in the direct measurements, the overall concentrations compare well. More will be done in some

upcoming research which will provide even better insights into sample stability. The bottom line answer to the question about stability is that the canisters we have used have appeared to work well during many previous campaigns, and the results from DRIVE look consistent. From this we conclude that sample stability over the storage time until analysis was not a significant issue. We include a little more detail into the manuscript in method section 2.1.

## 5. P19707 2.2 L18. Fixation of samples for flow cytometry may cause cell loss, particulary amongst nanophytoplankton, was this considered in the analyses?

We thank the reviewer for pointing this out. This has not been taken into consideration yet. To address this concern, a comparison between fresh (unfixed) and with GDA conserved samples is currently planned for the next campaigns. We include a corresponding reference in the methods section 2.2: *"Potential cell loss associated with the sample fixation has not been taken into account."* 

#### 6. P19710. 4.1.1 L5, Sentence starting 'While maximum. . ..,' requires rephrasing.

We rephrase this sentence to: "Maximum mean oceanic  $CH_3I$  of 3.0 (1.7 – 5.4) pmol  $L^{-1}$  was observed at S1. S3 showed the lowest mean  $CH_3I$  concentrations of 1.2 (0.2 – 2.1) pmol  $L^{-1}$  during 24 h."

## 7. P19710. 4.1.1 L14. Please explain how the difference in 'regional variability' between air and ocean CH<sub>3</sub>I concentrations was determined.

We have determined the variability as relative standard deviation from the overall mean between the individual stations and the measurements in between of each atmospheric and oceanic halocarbons. We state this clearer in the manuscript.

8. P19711 4.1.3. I would suggest presenting the results of the saturation anomalies and concentration gradients, fluxes and air-sea flux collectively for the 3 compounds. At present the separate sections involve a fair amount of duplication.

We thank the reviewer for the suggestion. We think that merging these two sections into a joint chapter might be a bit confusing. Our intention of presenting the results and the discussion each separate for  $CH_3I$  and the bromocarbons was to provide a better overview on the different results. However, we agree with the reviewer that there is a certain amount of duplication, which is why we look through the result chapters and reduce the replicated sentences to improve readability.

### 9. Fig. 4, it would make the figures clearer if the black and grey lines used, were more distinct.

We agree, and edit all the figures accordingly.

10. P19716, 5.1.1 L20+ Sources of CH<sub>3</sub>I. The conclusion that a lack of correlation between CH<sub>3</sub>I with Prochlorococcus, divynl chorophyll a, diagnostic diatom pigments and chlorophyll is not sufficient evidence to discount a biological source. For several reasons, i) correlations are a relatively weak means of determining a source, especially if the variables are being cycled at different rates ii) since both production and loss processes determine the observed concentrations; iii) there is evidence that heterotrophic bacterial processes may be involved in CH<sub>3</sub>I production (Manley and Dastoor 1988, Amachi et al. 2001, 2004, and others) and these correlations do not take account of these potential processes.

We thank the reviewer for this advice. We agree to all points, and take the reviewer's objections into account. We weaken our conclusion that we can completely rule out direct biological sources for oceanic CH<sub>3</sub>I in the investigation region. We realize that we have no information on the time scales of the processes that contribute to the formation or decomposition of CH<sub>3</sub>I during the campaign with the methods and the measurements we used. To shed light on these processes in the field, future campaigns should involve in-situ incubation studies that take both production and loss processes into account. We mention the limitations of our approach now with the following addition: "Lacking correlations of  $CH_3I$  concentrations with pigment and flow cytometry data does not necessarily allow for excluding a biological source completely. There may be a balance between potential production and loss processes, as well as involvement of heterotrophic bacteria during the campaign."

11. 19718, 5.1.2. Comparison to previous studies: The authors should at least try to explain the large difference in concentrations measured for  $CH_3I$ , between this study and that of Jones et al. 2010 and Richter and Wallace 2004? There is considerable debate at present regarding the relative contribution of different sources of reactive iodine to the tropical MABL and the 10-fold difference in concentrations between studies make this even harder to assess unless some explanation is provided.

We agree with the referee that the discrepancy in oceanic CH<sub>3</sub>I concentrations between the three studies is very large. The study of Richter and Wallace (2004) was conducted further south towards the equator. This region could be a stronger source region for oceanic CH<sub>3</sub>I as a result of photochemical production. In his dissertation, Richter (2004) explained the higher CH<sub>3</sub>I water concentrations in the region with low wind speeds that resulted into a smaller seato-air flux and hence a smaller sink for surface CH<sub>3</sub>I. Additionally, a relationship between high CH<sub>3</sub>I and high SST (higher than during our study) was found. We in contrast could not see an overall relationship of wind speed with sea surface concentrations of CH<sub>3</sub>I during DRIVE. However, we did measure the most elevated oceanic CH<sub>3</sub>I during the cruise at the diel station with the lowest wind speeds, supporting his hypothesis.

The study of Jones et al. (2010) covered the same region and the same season as our study, measuring up to 6 times higher sea water concentrations. We compared our oceanic CH<sub>3</sub>I concentrations with the climatology and the concentrations reported in the supplementary material of Ziska et al. (2013). The lower concentrations measured during DRIVE fall well within the general range of other oceanic regions, except for some shelf regions that are characterized by high concentrations possibly related to high macroalgal abundance. We conclude from this that the concentrations reported by Jones et al. (2010) are exceptionally high for an open ocean region. Unfortunately, there is only a limited amount of additional measurement parameters mentioned in the study, which makes it difficult to assess the causes for the difference to our study. Smythe-Wright et al. (2006) showed elevated concentrations of CH<sub>3</sub>I of up to 45 pmol L<sup>-1</sup> south of 40 °N which was attributed to increased *Prochlorococcus* abundance. Apart from photochemistry, this could be an explanation for the highly elevated CH<sub>3</sub>I concentrations during the study of Jones et al. (2010).

We add a condensed version of the potential reasons for the large discrepancies between the studies to the discussion section 5.1.1.

### 12. 19719, 5.2.1, L5. Measurements of algal abundance or pigments do not constitute 'algal activity', they may be a useful proxy for algal biomass.

We thank the reviewer for pointing this out. We reword the sentence as follows: "In contrast to oceanic  $CH_3I$  during DRIVE, oceanic  $CHBr_3$  and  $CH_2Br_2$  was elevated in the biological active regions and correlated with algal biomass parameters."

13. 19719, 5.2.1 Sources of bromocarbons. As for CH<sub>3</sub>I, the strength of correlation between observed concentrations and specific phytoplankton taxonomic groups is relatively weak evidence of a source. Firstly, attributing specific pigments concentrations to specific phytoplankton taxonomic groups is itself generally a weak proxy. For instance, how was diatom biomass determined in the present assessment? If it is based on the concentrations of fucoxanthin, then this compound is also found in the majority of haptophytes. The authors need to explain: i) how and which marker pigments were attributed to which phytoplankton groups; ii) make clear the uncertainty in their assessment of the link between these defined groups and the concentrations of the halocarbons; iii) point out that observed concentrations represent a balance between production and loss processes, e.g. a compound could exist at low concentrations but have both high production and high loss rates.

We acknowledge the reviewer's argument that correlations with phytoplankton pigments are not necessarily a clear indication for a source. However, previous studies could show that CHBr<sub>3</sub> and CH<sub>2</sub>Br<sub>2</sub> are often elevated in regions with increased phytoplankton abundance such as upwelling areas (Quack et al., 2004; Carpenter et al., 2009; Raimund et al., 2011) agreeing with the observations during DRIVE.

i. We have not directly determined biomass, but have linked significant pigments to phytoplankton groups which we have presented in Table 5. We agree that these links may not be as distinct. E.g. fucoxanthin to some extend is also present in other phytoplankton groups, thus we wrote "indicative" for a particular algae group. We state clearer that attributing pigments to certain phytoplankton groups is associated with uncertainties in section 5.2.1 of the manuscript: "Possible biological sources during DRIVE were identified by using phytoplankton pigments indicative for various phytoplankton groups which were investigated with MLR more thoroughly. However it should be noted that e.g. fucoxanthin occursg mainly in diatoms, but is also present in

other phytoplankton groups to a certain extent (Jeffrey and Vesk, 1997). The production of halocarbons and the occurrence of the phytoplankton pigments may also take place on different time scales, which may obscure or stimulate a correlation."

- ii. We state the uncertainties clearer and modify the sections (see also answer to i.).
- iii. We also agree with the reviewer with respect to the third point. Quack et al. (2007) found relationships between oceanic CHBr<sub>3</sub> and lower concentrated pigments showing the necessity to perform production-related studies rather than to conclude sources from single correlations. We have attempted to look more detailed into possible sources by applying a multi linear regression method with all pigment data with the exception of Chl *a*. However, we agree that it is difficult to determine the balance of and distinguish between production and loss processes with the methods we used. We clarify this in the manuscript similar to the statement we used for CH<sub>3</sub>I, and point out the limits of our approach. We agree that, similar to CH<sub>3</sub>I, in-situ incubation studies, along with measurements of primary production, could be a helpful instrument to shed light on these unknown factors.

14. 5.1 and 5.2 Discussion: Air-sea flux estimates and determination of the drivers: In a number of ways this paper demonstrates the need to now go beyond wind-based parameterizations of transfer velocity to estimate flux rates and to explore what really determines the flux. Several reports have recently demonstrated the value of using a more comprehensive, albeit more complex, algorithm to estimate and explore the controls on air-sea flux. The authors should, at least, address the limitations of their approach and explain what additional measurements would be required to implement an approach such as the COARE model (Fairall et al. 2003) for their flux estimates. [Fairall, C. W., E. F. Bradley, J. E. Hare, A. A. Grachev, and J. B. Edson (2003), Bulk parameterization of air to sea fluxes: Updates and verification for the COARE algorithm, J. Clim., 16, 571–591.]

We agree that a clear limitation of our approach is that we discuss the observations within the current available concept for the sea-to-air flux calculations involving wind speed and SST as main driving factors. The parameterization of the wind speed dependent compound specific transfer coefficient  $k_w$  does not consider stability of the atmosphere and the ocean, sea state, and additional surface processes such as surfactants that could possibly reduce the sea-to-air

flux, which may influence the transfer across the air-sea interface. In contrast, the COARE algorithm takes diffusive and turbulent mechanisms in the sea surface and the atmosphere influencing the gas transfer coefficients into account. This algorithm includes among others Vaisala air temperature and specific humidity, solar irradiance, downwelling longwave irradiance, and precipitation as additional parameters to wind speed, SST and air-sea concentration gradient. Apart from diffusivity and dissipation in the interfacial layers, the stability of the lower atmosphere, and wind stress according to the roughness of the sea surface, the available TOGA COARE program also includes bubble transfer. These are just some examples of data and variables that are included in the determination of the gas exchange coefficient using an algorithm such as COARE requiring a set of additional meteorological parameters. We add statements regarding the limitations of the used parameterization including the reviewer's suggested reference to the discussion chapter 5.1.2.

# 15. 5.4. 19722-3. Atmospheric lifetimes. The authors should explain whether their assessment of the contributions to atmospheric halocarbon mixing ratios takes into account losses within the atmosphere, and if not, why this is not required.

The average tropical atmospheric lifetimes reported by the WMO are 7 days for  $CH_3I$ , 24 days for  $CHBr_3$ , and 123 days for  $CH_2Br_2$  (Montzka and Reimann, 2011). Considering a fetch of 200 km and a mean wind speed of 7.4 m s<sup>-1</sup> (Fuhlbrügge et al., 2013), less than 3 % of the originally emitted  $CH_3I$  would have been removed which would be much less for  $CHBr_3$  and especially for  $CH_2Br_2$ . Since the influence of chemical loss processes in the atmosphere on this short time scale is very low, we decided for the sake of simplicity to neglect the chemical loss processes in the budget calculation. We add this explanation to the discussion section 5.4.1.

16. P19723 – 19726, 5.4.2. and 6. Discussion and Conclusion. The conclusion that MABL height has an influence on the air-sea flux of the bromocarbons is interesting but would benefit from some explanation of what may control that height and how that may change in the future, if at al.

The MABL over upwelling regions is mainly controlled by the difference between SST and surface air temperatures (SAT). Variations of the MABL height can be caused by SST variations on the one hand, especially due to changes of upwelling strength. However, Mote

and Mantua (2002) did not find any distinct changes in upwelling strength for the Mauritanian upwelling resulting from sea level pressure field changes until the end of the 21st century in their HadCM3 and CSM simulations. Different studies (e.g. Vecchi and Soden, 2007; Lu et al., 2007) reported an extension and weakening of zonal (e.g. Walker Cell) and meridional (e.g. Hadley Cell) cells. This would weaken the large scale surface wind fields and the surface wind stress, which is responsible for the upwelling and the stable boundary layer conditions as the warm air is transported offshore from the African continent. Nevertheless there are still uncertainties in the projections of future upwelling changes. The influence factors and the correlation of the VSLS to MABL height was extensively discussed in Fuhlbrügge et al. (2013) which we refer to in the manuscript.

# 17. Finally, the authors should also make it clear that these three compounds are only a subset of the suite of volatile halocarbons that may exchange between ocean and atmosphere and explain why they focus particularly on these specific compounds.

We agree with referee #1 that it is important to acknowledge the wide set of halocarbons that are transported from the ocean to the atmosphere. CHBr<sub>3</sub> and CH<sub>2</sub>Br<sub>2</sub> are usually among the most abundant halocarbons in the marine environment and represent the largest contributors to organic bromine from the ocean to the atmosphere (Hossaini et al., 2012). Although it is known that CH<sub>3</sub>I is the most abundant organoiodine in the atmosphere (Saiz-Lopez et al., 2012), there are still many uncertainties regarding the production pathways and the impact factors of sea-to-air fluxes of CH<sub>3</sub>I, which can be applied to CHBr<sub>3</sub> and CH<sub>2</sub>Br<sub>2</sub> as well. We express clearer in the manuscript the necessity to broaden the current knowledge involving these three halocarbons in the introduction.

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