<u>Response to comment by reviewer #1</u>

We deeply thank the reviewer for the effort invested in reviewing this paper and for its thorough and constructive review.

Interactive comment on "Emission of volatile halogenated organic compounds over various landforms at the Dead Sea" by Moshe Shechner et al.

Anonymous Referee #1
Received and published: 16 January 2019

1. *This is a study of concentrations of short-lived halogenated gases from a unique area, the Dead Sea. Given the interest in these chemicals and the uniqueness of this location, this paper has the potential to be an interesting contribution. It certainly includes a thorough review of the available literature and the authors have very thoroughly considered their new results in light of previously published work. However, I'm concerned about a number of aspects of the interpretation of the measurements, which are described below. The most significant is an inadequate consideration of uncertainties in most aspects of the work. This leads to an extended discussion throughout the paper of effects that I'm not convinced are real.*

In Table 2, comparisons are made between concentrations measured at these Dead Sea sites with reported concentrations in the marine boundary layer (MBL) (as medians, from Ozone Assessment Reports), and measured concentrations enhancements are taken to imply significant local emissions. But this seems an inappropriate conclusion. I would expect that the influence of the marine boundary layer on what is being measured in the Dead Sea valley is diminished by the time air moves from any distant sea (Red or Mediterranean) to this valley owing to vertical mixing within the lower atmosphere. Perhaps instead, any enhancement relative to the MBL suggests only that fluxes are non-zero in this region too, and are perhaps comparable (or larger) than suggested for the marine boundary layer and coastal ecosystems? Drawing conclusions from concentration differences in the Dead Sea area vs the MBL is tricky and not especially informative, given that concentrations are influenced by dynamics in addition to flux–this seems worth mentioning, but isn't yet in this regard. Also, why aren't MBL fluxes also shown in Figure 2?

<u>Answer</u>: Thank you for this comment. The only reason that we compared concentrations at the Dead Sea to those of the MBL was to suggest irregularly high local emissions in the Dead Sea area, rather than from either the Red Sea or the Mediterranean Sea, whose contributions to the local concentrations are indeed expected to be negligible. Considering the relatively large distance from the Mediterranean Sea (~90 km) and the Red Sea (160 km), we believe that these elevated concentrations imply local emissions from the Dead Sea area itself. We did not intend to indicate significant contributions from the Red or Mediterranean seas, and to clarify this, we now state the following: "Overall, the measurements at the Dead Sea boundary layer revealed higher mixing ratios for all investigated VHOCs than their expected levels at the Mediterranean Sea and Red Sea MBL, indicating higher local emissions from the Dead Sea area" (lines 333-335). We agree that comparing the mixing ratios at the Dead Sea to those measured at the MBL is tricky, but we think that it provides some understanding of how this area can contribute to VHOC loading relative to nearby marine environments. Taking this and the next comment into account, we now present the comparison of mixing ratios to those in the MBL in a figure without including enrichment factors.

We do not include MBL fluxes in Fig. 3 (originally Fig. 2) because reported fluxes in the MBL were measured under very different conditions, which also resulted in remarkably large differences in their magnitudes. For instance, fluxes of VSLSs have been found to be significantly different in magnitude over the coastal area, open ocean, shelf and upwelling (Carpenter et al., 2009). Moreover, we also compare the measured fluxes with those measured over various landscapes, such as bare soil, soil mixed with salt deposits, and vegetation, because in the case of fluxes, we find it more suitable to compare the reported fluxes more selectively in the text.

2. *I also find it very difficult to internalize the information given in Table 2 as presented. I'd recommend the presentation of these results, if retained, also (or instead) as a figure. Furthermore, I'd suggest that any enhancement factor should also consider the reported range in the marine boundary layer concentrations so that the reader can better understand the degree to which the Dead Sea region concentrations actually are anomalous (regardless of reason, flux or meteorology). In addition, for some of these gases there are some well documented temporal, seasonal, and latitudinal variations in MBL concentrations that aren't well considered by the "annual average for 2012". As a result, I suspect that some of the EF's (CH3Br, perhaps also CH3Cl, CHCl3, and CH2Br2) are not accurate representations.*

Answer: We agree, and the results presented in former Table 2 are now included as Figure 2 in the new manuscript. Table 2 is now presented in the Supplement, but instead of showing the enrichment factors, we explicitly show the reported measurements for the MBL as compiled by Carpenter et al. (2014). We agree that presenting enrichment factors is problematic because of the sensitivity of the concentrations to season, latitude, meteorological conditions, investigated area within the MBL, etc., particularly for the specified species (CH_3Br, etc.). Therefore, we do not include emission factors in the new version, and we think that presenting the results vs. the information compiled for the MBL in a graphical way, including a range for the MBL mixing ratios, is a reasonable way to compare the two data sets. Factors which may lead to biased comparison between mixing ratios measured at the Dead Sea and those measured in the MBL are now discussed: "It should be noted, however, that while Fig. 2 implies elevated VHOC emission from the Dead Sea, comparison of mean or median mixing ratios of VHOCs for the Dead Sea with those for the MBL is not straightforward, considering that VHOC mixing ratios in the MBL are sensitive to several factors, including season and latitude. Moreover, the measurement height can play a significant role in affecting the mixing ratios due to decreasing mixing ratios with height over areas where local emissions occur. Hence, we also compared the measured fluxes and mixing ratios with their corresponding values measured in coastal areas, where the highest mixing ratios in the MBL were generally measured due to stronger emissions." (lines 347-355). We also refer specifically to differences in sampling heights with respect to Fig. 5: “Note that differences in sampling heights at different sites can lead to a biased comparison between mixing ratios at different sites; nevertheless, in most cases, differences across measurement sites were larger than across vertical heights. “ (lines 592-594).

3. *On fluxes, the text seems to inaccurately reflect what the figure indicates once uncertainties are considered. One example: "Figure 2 presents the measured fluxes of all VHOCs studied. On average, the net fluxes of all measured species, except C2HCl3 and CH3I, were positive at most of the investigated sites", and my review of Figure 2 indicates a much lower occurrence of positive flux: only 13 of 36 panels (excluding CH3I and C2HCl3) show positive fluxes where the standard error does not encompass zero. Another example can be found in section 3.2.1, lines 417-419. It is necessary to consider the uncertainty on the average here in drawing conclusions. Furthermore,I would estimate that the standard errors are likely underestimated as a result of the fairly small number of measurements used to estimate fluxes in this work.*

Answer: We have addressed this comment by rigorously taking statistical significance into account throughout all of the analyses and discussions, and in drawing the related conclusions. Fluxes in Table 2 (originally Table 4) are presented along with *p*-values that indicate their statistical significance, for a specific species at a specific measurement site, by applying a one-sample t-test. Note that considering the small number of measurements, these *p*-values are presented in four different categories: $p < 0.05$, $0.05 < p < 0.1$, $0.1 < p < 0.15$ and $p > 0.15$. For our analyses, only *p*-values <0.05 are considered, indicating that a specific site is a net source or sink for a specific species, while the other two *p*-value categories (excluding $p > 0.15$) are used only to indicate a moderate likelihood of the fluxes being either positive or negative, possibly due to the small number of measurements. Note that in several cases, correlation analyses of both flux and mixing ratios strongly support the emission of species from a specific site, although the corresponding flux is reported as insignificant in Table 2; for instance, remarkably high correlations were found for CH_3I with $CHCl_3$ and C_2HCl_3 at the coastal sites near the seawater ($r = 0.99$, $p < 0.05$ in both cases), based on the flux correlation analysis (Sect. 3.3, lines 883-887), also supported by the concentration correlation analysis (Sect. 3 lines 887-895). Nevertheless, these coastal sites were found insignificant as a net source for $CHCl_3$ ($0.05 < p < 0.10$), and for this reason we think that presenting also 0.10 < *p*-values < 0.15 and 0.05 < *p*-values < 0.10 contributes, particularly for future studies in this field, even though these are not taken into account by our analyses.

Note that we also tested the *p*-value calculations assuming that EGD-SD and TKM-SD, as well as BARE–MSMR and BARE–MSD, are the same emission source, considering their similar characteristics (see Sect. 3.1). In the case of the SD sites, this assumption resulted in a lower evaluated *p*-value in only a few cases, affecting the *p*-value category (ranking; see Table 2). The statistical tests related to the information presented in Table 2 are described in its caption (see lines 495-497). This table was moved to Sect. 3.1 to support the reports on measured fluxes at the Dead Sea, based on the statistical analysis that is incorporated in the table. We have extensively changed the text in Sect. 3.1 and in Sect. 3.2.1–3.2.3 as a result of taking into account the uncertainties associated with the measured fluxes, based on the statistical analysis. In addition, the two specific comments given as an example by the reviewer were addressed (lines 419-421 and lines 549-552). Overall, however, the conclusions drawn based on the fluxes that are presented in Table 2 were not changed significantly. This is mainly because there is a general correlation between the incidence of positive fluxes associated with *p*-values $\leq$0.05 and that of the fluxes which were considered positive in the original version of the manuscript. We have double-checked the standard errors, provided by us as standard mean errors, and we find them to be accurate.

Regarding the correlation analyses, each correlation value (i.e., Pearson correlation coefficient (r)) in Tables 5 and 4, (originally Tables 6 and 5, respectively) is reported along with the corresponding *p*-value to indicate whether the correlation is significantly different from zero, based on Student's t-test. In the case of correlation coefficients, *p*-values are also reported in four different categories: $p < 0.05$, $p < 0.1$, $p < 0.15$ and $p > 0.15$. For the analyses, only *p*-values <0.05 were considered, indicating that a specific correlation is significant or not significant, respectively, while the other two *p*-value categories (excluding $p > 0.15$) were used only to indicate moderate likelihood of the fluxes being either positive or negative, taking into account the small number of available measurements.

The revisions resulting from taking into account the uncertainties for the analyses are included in sections 3.1–3.3, and in the following, we present the related revisions in the summary section, showing their overall effect on our findings and conclusions.

1. In the original version: "Overall, our measurements indicate a higher incidence (in 65−85 % of measurements) of positive fluxes of brominated than of chlorinated VHOCs, except for $CHCl_3$, for which the incidence of positive net fluxes was also relatively high (65 % of measurements)." (lines 756-759).

In the revised version: "Overall, our measurements indicate a generally elevated incidence of positive fluxes of brominated vs. chlorinated VHOCs compared to previous studies" (lines 910-911). Hence, this statement is valid based on the flux uncertainties, namely, considering a measured flux as positive only if the related measurement site was identified as a statistically significant ($p < 0.05$) source for the specific species (for which the flux was measured). In the revised version we do not specify the percentage of this positive flux, but the reader can find this information in Table 2, in several different statistical significance categories. We have removed the text on the incidence of $CHCl_3$ considering the relatively low incidence of positive flux from sites which were identified as a source for $CHCl_3$ ($p < 0.05$).

2. In the original version it was mentioned that: "The four investigated site types, the cultivated and natural vegetated, the bare soil and the coastal sites, are identified as potential net sources for all VHOCs investigated, except for the emission of CH_3I and C_2HCl_3 from the vegetated sites. Hence, this study reveals strong emission of VHOCs over at least a few kilometers from the Dead Sea" (lines 763-767).

In the revised version: "Three of the investigated site types – bare soil, coast and agricultural field – were identified as statistically significant ($p < 0.05$) sources for at least some of the investigated VHOCs. The fluxes, in general, were highly variable, showing changes between sampling periods, even for a specific species at a specific site. The coastal sites, particularly at a short distance from the sea (SD sites) where soil is mixed with salt deposits, were sources for all of the investigated VHOCs, but not statistically significantly for $CHCl_3$. Further from the coastal area, the bare soil sites were sources for $CHBrCl_2$, $CHBr_2Cl$, $CHCl_3$, and apparently also for CH_2Br_2 and

CH_3I, and the agricultural vegetation site was a source for $CHBr_3$, $CHBr_2Cl$ and $CHBrCl_2$. Our measurements reinforce reports of $CHCl_3$ and $CHBrCl_2$ emission from bare soil, but indicate that such emission can also occur under relatively low soil organic content. To the best of our knowledge, we report here for the first time strong emission of $CHBr_2Cl$ and emission of CH_2Br_2 from hypersaline bare soil, at least a few kilometers from the Dead Sea. We could not identify the contribution of either natural or agricultural vegetation to the emission of the investigated VHOCs." (lines 915-928).

3. In the original version: "Measurements at a bare soil site suggested a decrease in VHOC emission rates for 1−3 days after a rain event, while the gradual increase in VHOC emission more than three days after the rain event suggests that these VHOC emissions are, at least partially, biotic-induced." (lines 777-779).

In the revised version: "Rain events appeared to attenuate the emission rates of VHOCs at the Dead Sea. Measurements at a bare soil site suggested a decrease in VHOC emission rates for 1−3 days after a rain event." (lines 935-937). We do not include the hypothesis about biotic-induced VHOC emission, because it is less strongly supported if we consider measured flux as positive or negative only for measurement sites identified as statistically significant ($p < 0.05$) sinks or sources for the specific species (for which the flux was measured).

4. In the original version: "Trihalomethanes, including $CHCl_3$, $CHBr_2Cl$, $CHBr_3$ and particularly $CHBrCl_2$, are associated with the highest number of sites at which their flux was, on average, positive, while $CHBr_3$, $CHBr_2Cl$ and $CHBrCl_2$ showed relatively high incidence of positive fluxes, with values of 65 %, 80 % and 85 %, respectively." (lines 780-783).

In the revised version we do not include this sentence as is, because this cannot be supported, if uncertainties in a measurement site as source are taken into account. In the revised version, we focus more on the common mechanisms/controls for the emission of brominated trihalomethanes: "Both flux and mixing ratio correlation analyses pointed to common formation and emission mechanisms for $CHBr_2Cl$ and $CHBrCl_2$, in line with previous studies, for the agricultural watermelon-cultivation field and bare soil sites. These analyses further strongly suggest common formation and emission mechanisms for $CHBr_3$ with these two trihalomethanes." (lines 938-942).

5. In the original version: "The overall average net flux of the trihalomethanes decreased according to $CHBr_2Cl > CHBr_3 > CHBrCl_2 > CHCl_3$." (lines 790-791).

In the revised version we further support this point by using the flux magnitude: "The overall average net flux of the trihalomethanes decreased according to $CHBr_2Cl > CHCl_3 > CHBr_3 > CHBrCl_2$, while $CHCl_3$ showed the lowest incidence of positive fluxes among all trihalomethanes." (lines 944-946). Again, this finding of relatively elevated emission of brominated trihalomethanes (compared to previous studies) is generally supported by both the original and revised analyses.

6. In the original version: "We identified the SD sites as a probable source for all methyl halides, whereas vegetated sites appear more likely to act as a net sink for these species." (lines 796-797).

In the revised version: we realize that this sentence should be revised based on both the original and updated analyses: "We identified the coastal sites as a probable source for all methyl halides, whereas neither agricultural field nor natural vegetation site were identified as net sink or net source for these species, except for the agricultural field being a net sink for CH_3I." (lines 949-951)

7. In the original version: "Comparing the proportion of Br and Cl in the soil for the various sites with proportions of measured positive flux of CH_3Br and CH_3Cl are in line with reports by Keppler et al. (2001) about emission of methyl halides via abiotic oxidation of organic matter in the soil. Similar calculations in our study demonstrated much higher efficiencies of CH_3I emission than those reported by Keppler et al. (2000), pointing to emission of CH_3I via other mechanisms. The high correlation of CH_3I emission with that of $CHCl_3$ and C_2HCl_3, particularly at the SD sites, together with findings by Weissflog et al. (2005), of various chlorinated VHOCs emission, including $CHCl_3$ and C_2HCl_3, from salt lake sediments, suggests that the Dead Sea, particularly the SD, sites probably act as an emission source for $CHCl_3$, C_2HCl_3 and CH_3I via similar mechanisms. Weissflog et al. (2005) reported that the emission of chlorinated VHOCs in their study was induced by microbial activity. Keppler et al. (2000) reported the involvement of an abiotic process in the formation of alkyl from soil and sediments, and the observed correlation between methyl halides and both $CHCl_3$ and C_2HCl_3 may indicate that the two processes occur simultaneously." (lines 797-810).

In the revised version: The flux- and concentration-based correlation analyses in the revised version strongly support these findings, even though the coastal sites were not identified as statistically significant net sources for CHC_3l ($0.05 > p < 0.1$; see Table 2). The revised text refers to the statistical significance of the analyses and we have removed some experimental details about the related analyses to shorten the discussion: "Our analysis demonstrated, however, much higher efficiencies of CH_3I emission than of CH_3Br and CH_3Cl emissions as a function of halides in the soil, compared to those reported by Keppler et al. (2000), pointing to emission of CH_3I via other mechanisms. The strong correlation between both fluxes and mixing ratios of CH_3I, $CHCl_3$ and C_2HCl_3, particularly at the SD sites, strongly suggests that the coastal area of the Dead Sea acts as an emission source for $CHCl_3$, C_2HCl_3 and CH_3I via similar mechanisms, although these sites were associated with only moderate statistical significance ($p \leq 0.1$) as a net source for $CHCl_3$. The emission of $CHCl_3$ and C_2HCl_3 from these sites is in line with findings by Weissflog et al. (2005) of emission of various chlorinated VHOCs, including $CHCl_3$ and C_2HCl_3, from salt lake sediments. Weissflog et al. (2005) reported that the emission of chlorinated VHOCs in their study was induced by microbial activity. Keppler et al. (2000) reported the involvement of an abiotic process in the formation of alkyl from soil and sediments, and the observed correlation between methyl halides and between CH_3I and both $CHCl_3$ and C_2HCl_3 may indicate that the two processes occur simultaneously in the coastal area of the Dead Sea." (lines 951-965).

4. *Inferences about flux from measurements as a function of height have a certain spatial influence function. Please indicate what that might be for the sampling heights you have chosen.*

Answer: All measurement sites were carefully selected to ensure a sufficiently large homogeneous fetch, and the measurement height was chosen to ensure that the footprint falls within this homogeneous fetch, except for the SD sites where direct emission and uptake from the seawater can potentially affect the samples (lines 236-240). For EGD-SD-s and EGD-SD-w, the footprint included the seawater:" Based on the wind direction, in both cases, the sampling footprint included both the seawater and a narrow strip of bare soil mixed with salty beds (estimated at about 60% of the footprint) very close to the seawater." (lines 667-669). According to our calculations, the 80% footprint in the studied area ranged from ~100–950 m, which was, in all cases, significantly smaller than the fetch of any site (see Table S4). In some cases, the 90% footprint was ~2 km, but taking into account the wind direction for these specific cases (Table S6), the footprint was still smaller than the fetch, except for the samplings at the COAST–EGD, as described above. Based on this and the next comment, we realized that information was missing in the text about footprint and measurement height selections. Therefore, we have added the following text: "By default, the differences in height between the canisters increased exponentially with height, considering the typical decrease in the vertical gradient of emitted species in the surface layer (Stull 1988). All canisters were placed high enough above the ground to ensure that all sampling was performed within the inertial sublayer, except for the lowest canister at TMRX−ET. In all cases, the sample footprint fell inside the target fetch, except for the sampling at COAST–EGD, for which the sample footprint included a narrow strip of the seawater (estimated at about 40% of the footprint)." (lines 233-240).

5. *Consideration of the C2HCl3 results (an implied sink, perhaps from elevated mixing ratios in the broader Red Sea region) may indicate that the fluxes you are deriving here for naturally-emitted gases are actually not representative of the local regions you intended them to represent. How is the reader to assess this? Also, what has determined the different heights at which samples were collected on these masts? Sampling heights in a region with local emissions should have a large, but not discussed, impact on measured mixing ratios–which are being compared among sites and to MBL results.*

Answer: Our analyses suggest that in general, the investigated sites act more like a sink for C_2HCl_3, with the coastal sites probably also being a source for this species, based on the mixing ratios and flux correlation analyses (Tables 4 and 5), and the measured positive fluxes from these sites (Table 2). We cannot rule out that the sink for this species reflects its emission from the Red Sea, the Mediterranean Sea, or an anthropogenic source upwind. However, it is not likely that emission from the Red or Mediterranean Sea impacts the measured fluxes at the Dead Sea due to the following: (i) we added correlation and wind direction analyses, including for C_2Cl_4, which strongly support the origin of C_2HCl_3 from an anthropogenic source (see Sect. S4); (ii) the Red Sea and the Mediterranean Sea are located 160 km and 90 km from the Dead Sea, respectively, while mixing ratios of the investigated VHOCs at the Dead Sea are typically significantly higher than those in the MBL, and therefore probably also compared to those over the Red Sea and Mediterranean Sea; (iii) prevailing wind direction during the (different) measurement periods was from the north and in only a few cases, from the northwest (Table S6), whereas the Red Sea and Mediterranean Sea are located to the south and west of the Dead Sea, respectively; (iv) there is no reason to assume that the Dead Sea is an efficient sink for VHOCs transported from the Red Sea and the Mediterranean Sea, whereas there are efficient sinks for these species along the trajectories of the air masses. Therefore, it is not likely that the Dead Sea acts as a significant sink for VHOCs which are transported from these seas.

The Dead Sea probably acts more as a sink than a source for C_2HCl_3, but based on the above, this is more likely be the result of emission from inland anthropogenic sources in Israel. In any case, deposition of C_2HCl_3 or any other species cannot contribute to the emission fluxes at the Dead Sea—the latter, and not deposition, being the focus of this study.

Pursuant to this comment, we include the following: "Only COAST–EGD and COAST-TKM-SD sites were found to be statistically significant sources ($p < 0.05$, see Table 2) for C_2HCl_3, suggesting that the elevated mixing ratios for this species in the Dead Sea area result mostly from local anthropogenic emissions. This possibility is supported by the high correlations with C_2Cl_4 (Table S5). Emissions from a more distant natural source, such as the Mediterranean Sea or Red Sea, are unlikely given their large distance away (~90 km and ~160 km, respectively)." (lines 488-493).

To address the comment about sampling heights, we include the following: "By default, the differences in height between the canisters increased exponentially with height, considering the typical decrease in the vertical gradient of emitted species in the surface layer (Stull 1988). All canisters were placed high enough above the ground to ensure that all sampling was performed within the inertial sublayer, except for the lowest canister at TMRX−ET. In all cases, the sample footprint fell inside the target fetch, except for the sampling at COAST–EGD, for which the sample footprint included a narrow strip of the seawater (estimated at about 40% of the footprint)." (lines 233-240).

In the revised version, we discuss the impact of measurement height as well as of additional factors, including season and latitude, on differences in mixing ratios between our study and the MBL: "It should be noted, however, that while Fig. 2 implies elevated VHOC emission from the Dead Sea, comparison of mean or median mixing ratios of VHOCs for the Dead Sea with those for the MBL is not straightforward, considering that VHOC mixing ratios in the MBL are sensitive to several factors, including season and latitude. Moreover, the measurement height can play a significant role in affecting the mixing ratios due to decreasing mixing ratios with height over areas where local emissions occur. Hence, we also compared the measured fluxes and mixing ratios with their corresponding values measured in coastal areas, where the highest mixing ratios in the MBL were generally measured due to stronger emissions." (lines 347-355).

6. *I find the results in Figure 3 intriguing, although not much is made of it in the text. While it may be that no generalizations are possible related to all gases, there are some interesting similarities that might be worth discussing, especially to understand if these co-variations are consistent with the discussions related to co-variations in fluxes as what was intended in Table 4.*

Answer: First, note that Fig. 5 (originally Fig. 3) has been revised according to a comment made by reviewer #2 (we removed the data for VHOCs that did not show any seasonal variation), and the original figure is presented as Fig. S1 in the Supplement. The only case for which we suspect that there was a clear seasonal effect on VHOC mixing ratios is CH_3I, where both flux and mixing ratio measurements clearly indicated higher emission in spring vs. winter, apparently in line with findings by Sive et al. (2007), and accordingly we have added the following: "While no clear impact of season on mixing ratios was observed, for most sites, differences between two measurement sets resulted in consistent differences in mixing ratios, such that one measurement set resulted in higher mixing ratios for all or most species than the other. This suggests that other factors play a significant role in emission rates of all or most VHOCs in the studied area. Only the CH_3I results indicated moderate statistical significance ($0.05 < p < 0.1$) for higher mixing ratios in the spring vs. winter, in agreement with seasonal trends for its flux, as discussed above." (lines 598-604).

We further add, in Sect. 3.2.3 on lines 763-766:" As discussed in Sect. 3.2.1, the mixing ratios of CH_3I also tended to be higher in magnitude in spring compared to winter, with moderate statistical significance ($0.05 < p < 0.1$ in both cases) (Figs. 3, 5)."

Considering this comment, we have modified Table 4 (originally Table 5), and the correlations between mixing ratios are now provided individually for different site types, similar to Table 5 (originally Table 6), and also for all site types except for SEA–KDM (this site explores the effect of air transported over the seawater on the mixing ratios). These analyses indeed enabled us to further strongly support apparent common emission sources and/or controls between brominated trihalomethanes (Sect. 3.3, lines 846-882) and between $CHCl_3$, CH_2Cl_3 and CH_3I (Sect. 3.3, lines 883-895), as well as between methyl halides (Sect. 3.3, lines 883-886).

7. *Table 4 is also very hard to extract information from... and as before, I'm concerned that any identification of positive flux amounts don't take into account uncertainties on those estimations. If uncertainties were not considered, then it seems that much of the discussion related to incidences of positive flux and rankings by chemical etc. that follows should be reconsidered.*

Answer:

We have realized that the information on the ratios between flux and mixing ratio (defined as F:C in the original version) does not contribute significantly to the manuscript, and based on this comment, we have removed the F:C information from the table (now Table 2). We believe that this makes the table easier to extract data from.

As explained above, we include in the revised manuscript *p*-values, indicating the statistical significance of the related measurement site being a source for a particular species. Only *p*-values ≤ 0.05 were considered statistically significant in the analyses in the revised manuscript. The resulting differences in our conclusions and findings are summarized in our response to comment #3. Overall, they were not significantly changed by taking into account statistical significance.

8. *Table 5 and 6 need a consideration of correlations that are and are not significant, given the number of measurements included in each determination. Given the small number of samples considered here, I'd estimate that correlations of <0.1 are in fact indicative of no evidence for a correlation, not a correlation described as "low".*

Answer: We agree. All correlations in Tables 4 and 5 (originally Tables 5 and 6) are now reported with a corresponding *p*-value, based on Student's t-test, to indicate whether the correlation is significantly different from zero. Similar to the revisions we made to Table 2, we consider four different categories of *p*-values: $p < 0.05$, $p < 0.1$, *p* < 0.15 and *p* > 0.15. Only *p*-values <0.05 are considered statistically significant in the analyses in the revised manuscript.

References

Carpenter, L. J., Jones, C. E., Dunk, R. M., Hornsby, K. E., and Woeltjen, J.: Air-sea fluxes of biogenic bromine from the tropical and North Atlantic Ocean, Atmos Chem Phys, 9, 1805-1816, 2009.

Carpenter, L. J., Reimann, S., Burkholder, J. B., Clerbaux, C., Hall, B. D., Hossaini, R., Laube, J. C., and Yvon-Lewis, S. A.: Ozone-depleting substances (ODSs) and other gases of interest to the Montreal Protocol, Chapter 1 in Scientific Assessment of Ozone Depletion: 2014, Global Ozone Research and Monitoring Project – Report No. 55, World Meteorological Organization, Geneva, Switzerland, 2014.

Keppler, F., Eiden, R., Niedan, V., Pracht, J., and Scholer, H. F.: Halocarbons produced by natural oxidation processes during degradation of organic matter, Nature, 403, 298-301, Doi 10.1038/35002055, 2000.

Keppler, F., Eiden, R., Niedan, V., Pracht, J., and Scholer, H. F.: Halocarbons produced by natural oxidation processes during degradation of organic matter (vol 403, pg 298, 2000), Nature, 409, 382-+, DOI 10.1038/35053144z, 2001.

Stull , R. B.: An introduction to boundary layer meteorology. Kluwer, Dordrecht 1988.

Weissflog, L., Lange, C. A., Pfennigsdorff, A., Kotte, K., Elansky, N., Lisitzyna, L., Putz, E., and Krueger, G.: Sediments of salt lakes as a new source of volatile highly chlorinated C1/C2 hydrocarbons, Geophys Res Lett, 32, Artn L0140110.1029/2004gl020807, 2005.