

Response to comment by reviewer #2

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

General Comments

The manuscript by Shechner et al. presents ambient measurements and fluxes for short-lived halocarbons at multiple sites around the Dead Sea. The unique characteristics of the Dead Sea make it a very interesting location to study the emissions from and detail the characteristics of this source for atmospherically important halocarbons. The paper contains an abundance of information, but I feel some key details are lacking that are needed to fully assess the author's interpretations. Additionally, the paper is very long and becomes difficult to follow in terms of the main points trying to be conveyed in the various sections of the paper. My opinion is that the paper could be distilled down in length and the key points be fleshed out a bit more cleanly. Additionally, I feel there are some significant improvements that could be made in dissemination of the information in both graphical and tabular form. While there is merit to the manuscript, I feel as though there are an array of issues that should be addressed before it is in an acceptable format for publication.

I will present a general list of issues here and elaborate on them in the Specific Comments section.

Urban and other source influences – it would be useful to provide some context to the potential of urban emissions, for the solvents like CHCl_3 and C_2HCl_3 , but also including things like wastewater treatment facilities and other agricultural activities that could influence the area.

The first time a chemical constituent is introduced, it should be spelled out – there are several places this occurs throughout the manuscript. For example, L72 chloroform (CHCl_3), L73 chloroethane ($\text{C}_2\text{H}_5\text{Cl}$), L112 iron (Fe), L115 potassium bromide (KBr), nitric acid (HNO_3), etc. – please address.

Percentages – there are spaces between the number and the percent sign. The most common convention is to not have a space between a number and the percent sign.

I would recommend either referring to the suite of halocarbons as VHOCs or VSLS, but not going back and forth between them.

From the measured fluxes, can you estimate the local/regional source or sink strength of the Dead Sea? How do your results play in to the scale of the source strength of the Dead Sea for these gases?

It would be useful to present some quantitative information in the abstract, such as mixing ratios and fluxes.

*There are no uncertainties propagated through any of the fluxes.
I would be useful to include the atmospheric lifetimes and primary removal sources for the compounds in the manuscript.*

*The manuscript seem to try and agree with all previous studies.
Tables are difficult to read and digest.
Plots within the figures are too small making it difficult to extract information from them.*

Flux section could be moved to SI

A more thorough overview of the site, including meteorology, would be useful to help set the stage for the reader.

Answer: We have addressed all of the above according to the specific comments below and have revised the discussion to make it more concise. We have also added information in Sect. 2.1.1 regarding anthropogenic emission sources in the studied area, including agricultural fields, and mention that to the best of our knowledge, there are no wastewater facilities in the area of the Dead Sea. The reader is further referred to a sensitivity analysis which investigates, as suggested below, the potential impact of anthropogenic activity on the measured mixing ratios, based on the ratio $[C_2HCl_3]/[C_2Cl_4]$ (see Sect. S4). We further include a description of the meteorological conditions in the area of the Dead Sea (Sect. 2.1.1), and a summary of the meteorological measurements in Sect. S6.

In the revised version, we rigorously take into account the statistical uncertainties associated with the measured fluxes (and correlations) for our discussion and analyses, allowing us to present a clearer description of net emission/sink for the different species at the various investigated sites. Nevertheless, we do not yet have sufficient measurements to provide a reliable estimate of the overall emission/sink status for the various species at the Dead Sea. The Dead Sea area includes relatively highly diverse and changing landscapes, partly due to rapid evaporation, which leads to exposure of new deposits, and our measurements indicate high variability in emission, even for the same landscape. Therefore, many more measurements are required for a reliable estimate of total emission/deposition of VHOCs in this area. We have added flux and mixing ratio ranges for all species to the abstract and give lifetimes and primary removal pathways in Sect. S1, with a reference in the Introduction (line 84). We have revised original Figs. 2 and 3 and Table 2 for easier extraction of information, as suggested in the reviewer's comment below, and have included a new figure which provides a schematic of the flux's spatial distribution in the studied area.

Specific Comments

L46-7: You should include why CH3I and C2HCl3 are exceptions, as this is not intuitive to the reader.

Answer: The text in this case has been modified to account for statistical uncertainties associated with a measurement site being a source for the tested VHOC: "Fluxes were generally positive (emission into the atmosphere), corresponding to elevated mixing ratios, but were highly variable... Taking into account statistical uncertainties, the

coastal sites (particularly those where soil is mixed with salt deposits) were identified as the source for all VHOCs, but this was not statistically significant for CHCl_3 . Further away from the coastal area, the bare soil sites were sources for CHBr_2Cl , CHBr_2Cl , CHCl_3 , and probably also for CH_2Br_2 and CH_3I , and the agricultural sites were sources for CHBr_3 , CHBr_2Cl and CHBrCl_2 ." (lines 45-58).

L49-51: For the statement: "Correlation analysis, in agreement with recent studies, indicated common controls for the formation and emission of all the above trihalomethanes but also for CH_2Br_2 .", I'm not convinced this is entirely accurate – for example, what about CHCl_3 ? Also how does the correlation indicate that the factors controlling the formation and emissions are the same?

Answer: Two sentences have been included to address these two points based on the revised analyses which take into account statistical uncertainties: "Correlation analysis, in agreement with recent studies, indicated common controls for the emission of CHBr_2Cl and CHBrCl_2 , and likely also for CHBr_3 . There were no indications for correlation of the brominated trihalomethanes with CHCl_3 ." (lines 61-64). We agree that the correlation is not necessarily indicative of similar formation controls, but we believe that it is indicative of similar emission controls.

L55: "elevate" should be "elevated"

Answer: Corrected (line 68)

L61: When you introduce VSLs here, you should include here that this refers to compounds that have lifetimes of less than 6 months.

Answer: This is now included (line 75).

L64: replace "destruction of ozone" with "ozone destruction"

Answer: Changed accordingly (line 78)

L73: add "which", so it reads "... $\text{C}_2\text{H}_5\text{Cl}$, which originate..."

Answer: Amended (lines 87-88)

L134-5: Bromide (Br^-) and chloride (Cl^-) should be introduced and the sentence should be revised to read "with water salinity 12 times higher and a bromide to chloride ratio (Br^-/Cl^-) 7.5 times higher than in normal ocean waters.

Answer: Amended (lines 151-153)

L136: What do you mean by "landforms"? Formations from the residual salts left behind? In this case the use of the term "landform" invokes images of large scale topographical features, is this the case?

This brings in to question the use of the term landform in the title – is this really appropriate and accurate? I would say this work has been carried out on different terrains or ecosystems of the Dead Sea, but not different landforms.

Answer: We have replaced the term landform with landscape (including the title) to indicate that the entire ecosystem is being addressed, and not just geological formations. It is true that some of the measurement sites represent similar landscapes, as in the case of different distances from the seawater in the same area (e.g., for COAST-TKM-SD, COAST-TKM-LD), but in most cases, they differ fundamentally in some aspect (e.g., soil mixed with deposits at the coast vs. bare soil far from the

seawater vs. cultivated vegetation vs. natural vegetation) and the areas with the same characteristics are quite large (at least one to a few kilometers). We think that using "terrains" may imply different slopes, structures, etc. We replaced "landforms" on (original) line 136 with "newly exposed sea deposits" (line 154) to better describe the location.

L143: I would revise this to make it a stronger statement, something like: "Studying the emission of VHOCs at the Dead Sea is also fundamental for understanding local surface ozone depletion events..."

Answer: Thank you. We have modified the sentence accordingly (lines 161-164).

L169-70: Regarding the Tamarix vegetation and watermelon fields, more details, such as density, proximity, size of agricultural development, etc., would be useful to the reader.

Also, I would refer change your referencing of watermelon fields from "vegetation" to "agriculture" in later sections of the manuscript – because this is a perturbed system different that the natural vegetation, it should be distinguished as such.

Answer: More information is now included for the two sites, such as size, height, vegetation cover fraction and distance from the sea. In addition, meteorological conditions and estimated footprints are now included in the Supplementary Information (Tables S6 and S4, respectively). We refer in the revised manuscript to the watermelon field as an agricultural vegetation or agricultural field throughout the text. Note, however, that for the analyses, we refer to the natural vegetation and agricultural vegetation both as separate sites and as two vegetated sites (VEG; e.g., Tables 5 and 6).

L198: Revise to "Lastly, WM-KLY..."

Answer: Done (line 224)

P8, Sect. 2.1.2. How many samples were collected in total, at each site, and at each corresponding height for each site? What were the meteorological conditions during the sampling?

In order to get better feel for the results presented, both for the ambient levels and the fluxes, knowing N is critically important. This will allow the reader better perspective on some of the interpretation presented.

Also, general information about the seasonal and local meteorology to provide an overview of the region would be instructive to the reader.

Answer: In the revised version, sample information is specified in Table 1 and the number of samples at each site is presented in Fig. 2 (replacing original Table 2), while Table 2 (originally Table 4) and Fig. 3 present each evaluated flux individually; we specify that in two cases, flux was evaluated based on two samplings rather than three. Tables 4 and 5 (which present the correlation analyses; originally Tables 5 and 6) present the number of values used for the mixing ratio and flux correlations, respectively. We have added to the Supplement a table that summarizes the collected meteorological data during the different measurements (Table S6) and refer to it in the text (line 246). In the revised version, we include information about the climate of the

Dead Sea, including evaporation rate, annual precipitation and seasonal variation in daily maximum temperature (lines 177-181).

L209: Regarding the use of “fast” here: I personally wouldn't consider 20 minutes to be fast - I think the key point you are trying to make is that all samples were collected simultaneously and integrated over a 20 min period – please revise.

Also, I'm assuming “lifting of the canisters” should be “filling of canisters”

Answer: Thank you. The sentence has been revised to clarify this point: "To minimize non-synchronized air sampling by the three canisters, we constructed a special sampling system that allows almost simultaneous filling of the canisters" (lines 240-241).

L209-12: Please revise the following sentence – very awkward as written:

Facilitated by passive grab samplers (RESTEK Corporation, PA, U.S.), we performed each sampling within 20 minutes by pulling air into evacuated 1.9 L stainless steel canisters, resulting in an internal canister pressure higher than 600 torr.

Answer: The sentence has been revised: "For each sample, air was drawn into a 1.9 L stainless-steel canister via passive grab samplers (Restek Corporation, PA, USA), resulting in a sampling duration of 20 min and internal canister pressures higher than 600 Torr." (lines 242-244).

L215-16: Please revise: “...subjected to the analytical techniques...” – simply say they were analyzed by similar techniques described in Colman et al.

Answer: Amended (lines 246-248)

L218-21: You introduce all of the halocarbons here, but most, if not all should have been introduced previously. Please address.

Answer: Done

L223-28: Please provide some statistical/quantitative rationale for this - you can't simply disregard this point because it doesn't “agree” with the other measured mixing ratios for CH₃Cl. Also, I don't feel it's appropriate to state that it may result in a “less accurate flux” – how do we know what the “accurate flux” is? There is variability in all of this work, and while this may, in fact, be a spurious data point, what measures were carried out to deduce this issue?

Where is this listed, in Table 2? Please specify here for the reader to address.

Answer: This is now supported using Grubbs's test (Grubbs and Beck, 1972) and the text was revised according the comment (Sect. 2.1.2, lines 252-255). We now indicate this issue in Table 1 as well, and refer the reader to Sect. 2.1.2 (where this issue is described) from all relevant figures and tables.

L229, Table 1: It would be useful to provide the total number of samples and how many per height. This should be summarized such that the reader doesn't have to try and count how many samples were collected on the individual days from the information in the table.

Answer: We now specify the total number of samples for each experiment in Table 1. Note that the number of samples corresponding with each of the correlation analyses is now specified in both Tables 4 and 5 (which present the correlation analyses; originally Tables 5 and Table 6) as well as Fig. 2 (replacing original Table 2).

L279: Sect 2.3

Following suit with the canisters, how many total soil samples were collected and analyzed? This potentially could be moved to the SI because the information is only used for general properties at each site.

Answer: We mention now in Table 3 that: "Analyses were performed for a single mixture of samples at each site." (lines 693-694). This was of particular importance for the watermelon field site, where the area was clearly not homogeneous. A different number of samples for each site was used for each sampled mixture. We prefer to keep the table in the main text because we use some of the information presented there in our discussion, mostly in Sect. 3.2.2, regarding the content of halides (e.g., lines 652-664) and organic matter (e.g., lines 769-780).

L280-81: Please elaborate what you mean by this and what is the significance of this statement: "...at least 3 months following any rain event in the Dead Sea area."

Answer: This is to ensure that sample composition and water content are not affected by drifts and percolation, following recent rain events in the area, as is now explained in the text: "Soil samples at each site were collected up to a depth of 5 cm during the summer, at least 3 months after any rain event in the Dead Sea area, to ensure no impact on the samples by recent drift and percolation." (lines 311-313).

L265: In line reference should be Golder (1972)

Answer: Corrected (line 297)

L290: Quotes are not needed around "Discover"

Answer: Done (line 322)

L292-93: High Resolution does not need to be capitalized

Answer: Done (line 324)

L294: "low-limit" should be "lower limit"

Answer: Amended (line 326)

L302: What is meant by "corresponding available information."

Answer: We have removed this sentence and refer first to the measured mixing ratios: "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" (line 333-335).

L306: What is the "Dead Sea Works"?

Answer: A short description of the Dead Sea Works has been added: "The main anthropogenic emission source in the area, apart from local transportation and a few small settlements, is the Dead Sea Works, a potash plant located to the south of most of the measurement sites (see Fig. 1)." (lines 184-186).

L326: There were surface seawater, ambient air and direct flux measurements of CHBr₃ in Zhou et al., 2005 – how do these compare with the Dead Sea?

Answer: Information about mixing ratios of CHBr₃ as well as fluxes for CH₂Br₂ and CH₂Br₂ based on Zhou et al. (2005) has been added on lines 428-434.

L330: It appears that a range of values is missing after 2-60 pptv – there is simple “(-)”

Answer: The range has been added (line 373).

L335: Re C₂HCl₃ and CH₃I - while the reader can look at the figure, it would be useful to also state in the text what these gases are doing, on average.

Answer: This sentence has been deleted because it is not valid when statistical uncertainties are taken into account, which is the case in the revised manuscript. We now address these species as follows: "It can be seen that for all species, at least one of the six studied areas could be classified as a net source, with somewhat less sites being statistically significant net sources for CHCl₃, C₂HCl₃ and CH₃I. Note that as explained above, C₂HCl₃ was found to be affected by anthropogenic emission, which could explain the relatively less frequent identified emissions for this species "(lines 419-423). We also discuss the measured fluxes of CH₃I throughout Sect. 3.1, and compare them to fluxes reported in the literature: "The positive fluxes measured at BARE–MSMR were similar to the measured soil-emission fluxes of CH₃I reported by Sive et al. (2007) at Duke Forest, averaging ~0.27 nmol m⁻² d⁻¹ (range, ~ 0.11–4.1 nmol m⁻² d⁻¹)." (lines 485-487).

L336-39: Can you please clarify these two sentences: Figure 2 doesn't show values higher than these. Either present the values or revise text.

Answer: The sentence has been revised: "The flux magnitudes for CHBr₃ and CH₂Br₂ were greater than for most reported emissions in the MBL (e.g., CHBr₃, 25.2–62.88 nmol m⁻² d⁻¹ for the Mauritanian upwelling (Quack et al., 2007); CH₂Br₂, 0.14–0.29 nmol m⁻² d⁻¹ for the New Hampshire coast (Zhou et al., 2008)), but were smaller than the corresponding average fluxes estimated by Butler et al. (2007) for global coastal areas (~220 and 110 nmol m⁻² d⁻¹, respectively) and than the average flux from the New Hampshire coast as reported by Zhou et al. (2005) (~620 ± 1370 nmol m⁻² d⁻¹ and 113 ± 130 nmol m⁻² d⁻¹, respectively)." (lines 428-434).

L376: for the following, you either have one too many or one too few brackets: (e.g., ~600 nmol m⁻² d⁻¹; (Deventer et al., 2018).

Answer: Corrected (line 473)

L360: For “nmol m⁻²-d⁻¹” there appears to be an extra dash in between m-2 and d-1

Answer: Corrected (line 456)

L391-93: It is difficult to see this in Fig 2, and what/where are the anthropogenic emissions located? From DSW or other places? Can this be assessed by looking at something like the C₂HCl₃/C₂Cl₄ ratio? It is likely that this data is available from the UCI group, but this (or other pairs of compounds) could be used to do a more thorough analysis on the impact of anthropogenic emissions at the sampling sites. For example, this brings in to light things like wastewater treatment facilities and the corresponding emissions of CHCl₃ and CHBr₃. It would be useful to provide a more rigorous assessment of the influence of anthropogenic emissions in general - particularly for those not familiar with the region and to what extent they may be influencing this work - if minimal, that's great - just demonstrate this, as this

statement affects your results - C₂HCl₃ isn't the only gas here with anthropogenic sources.

Answer: We have added more details to the Methods about potential anthropogenic emission sources in the area (lines 184-190), and a thorough analysis of potential anthropogenic emission sources in general and particularly for C₂HCl₃ (Sect. S5). We summarize this analysis in the main text: "No association was observed between the measured mixing ratios and the air masses flowing from the direction of the Dead Sea Works (see Sect. S4 for anthropogenic impact), a potash plant located to the northwest of the TMRX–ET site and to the south of all other measurement sites (see Fig. 1) that is the main anthropogenic source in the area under investigation. Furthermore, the correlation analysis (Table S5) revealed that only C₂HCl₃ was associated with C₂Cl₄, a well-known anthropogenic VHOC. The absence of any other associations suggested dominance of natural sources for the VHOCs in the studied area." (lines 335-342). To the best of our knowledge, no wastewater facilities are located near the Dead Sea water.

The sentences have been revised: "Only COAST–EGD and COAST-TKM-SD sites were found to be statistically significant sources ($p < 0.05$, see Table 2) for C₂HCl₃, 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 C₂Cl₄ (Table S5)." (lines 488-491).

Suggestion: After looking at Figure 2, I feel as though it would be useful to have a summary flux figure (e.g., by compound) with the magnitude of the fluxes plotted by size or color on a map to enable the reader to get a better idea of the spatial variability of the flux magnitudes.

Answer:

Thank you. We have produced a new figure—Fig. 4—which focuses on the spatial distribution and variation in measured net fluxes at the various site types. The figure clearly demonstrates higher emission from the coastal area, particularly for the sites which are closer to the seawater and from the natural vegetation for some of the VHOCs, which are generally higher than for the cultivated field. However, the natural vegetation site could not be classified as a statistically significant source for the investigated VHOCs, pointing to the need for additional measurements at this site (lines 555-560). We also keep Fig. 2, which is focused on the effect of season and distance from the seawater.

L418: Revise to: "...VHOCs, except C₂HCl₃, were..."

Answer: The sentence has been revised: "The results presented in Sect. 3.1 showed elevated mixing ratios and net fluxes for all investigated VHOCs, with relatively less frequent positive fluxes for CH₃I, CHCl₃ and C₂HCl₃." (lines 547-548).

L431-33: Regarding the statement that there isn't a difference between fluxes in the spring and winter, two things should be addressed: 1) is this statistically significant? 2) What is the seasonality of the temperature and overall meteorology for this area (i.e., local/regional transport patterns)? Being only slightly extratropical, would seasonality be expected to be an important driver?

Answer: These two points have been addressed as follows: "Differences in VHOC emissions between winter and spring may arise from the generally much higher

temperature, and lower precipitation during the latter; further considering the high evaporation rate in this area, the soil water content is expected to be generally lower in spring compared to winter (Sect. 2.1.1; see also Table S6). Figure 3 suggests that there were no clear differences in VHOC fluxes between spring and winter, as supported by statistical analysis, except for CH₃I and CH₂Br₂ for which fluxes were higher in the spring, with moderate statistical significance ($0.05 < p < 0.1$).” (lines 578-584).

Seasonal variations between spring and winter in this area are relatively significant (e.g., summer mean daily maximal temperature of ~40 °C decreasing gradually until winter, to a corresponding temperature of ~21 °C; Sect. 2.1.1). Basically, the area is controlled by 19 main synoptic scenarios, with a dominant influence of local sea and Mediterranean breezes (Shafir and Alpert, 2011). In the manuscript, we focus directly on the meteorological parameters (which are further summarized in Table S6 for the measurement periods), and soil water content (which was not measured for the specific measurement periods), rather than the dynamics and synoptics that control these parameters.

L437: add comma after “properties”

Answer: Done (line 587)

L439-40: Regarding the sentence: “No clear impact of season or distance from the seawater on the mixing ratios can be discerned in this figure,...”, while I agree, it’s mostly because you can’t see the details in Figure 3.

Figure 3: In general, it is difficult to discern the spatial distributions and get useful information out of the vertical profiles because each panel is so small. From this figure, it is difficult to see and discern the gradients for many of the gases. I would recommend revising and either show a few key species and put the remainder that don’t show anything in the SI or revise the whole figure.

Answer: We have revised the figure as suggested (Fig. 5 in the revised manuscript), by excluding information for those species that did not show any seasonal trend. This indeed enables us to present a less "busy" figure with somewhat larger panels. We include the original figure in the Supplement (Sect. S2). Note that the aim of this figure is to explore the effects of season and distance from the seawater on the measured mixing ratios and therefore, we use a similar scale for the y-axis associated with the same species in the same coastal area (e.g., COAST–TKM-SD and COAST–TKM-LD). As a result, in many cases, it is still difficult to see or discern the gradients in mixing ratios due to the substantial range. Pursuant to this comment, we now indicate in the caption of Fig. 5 that the figure uses even y-axis scaling for sites in the same coastal area (line 615). To make it clear that the figure focuses on mixing ratio differences vs. distance, we have arranged the panels differently and schematically indicate the distance from the seawater by arrows.

L460: replace “these parameters” with “the soil composition parameters”; also change “The table records...” to “The results presented in Table 3 show...”

Answer: Amended (lines 627-628)

L462: “larger distance” should be replaced with “greater distances”

Answer: Amended (lines 629-630)

L465-66: replace “in” with “at” just before the site location abbreviation.

Answer: Done (lines 633)

L472-75: What do you mean by “underestimated value of Fe”? A lower limit of the total iron? Again, “low-limit” should be “lower limit”.

Answer: Yes, we mean a lower limit of the total iron. The sentence has been changed accordingly (line 639).

What does “while the emission rates became saturated” mean – I’m assuming that you mean “plateau”. For example, Huber et al. use the term “plateau”.

Answer: Yes, the sentence has been clarified: "Note, however, that soil Fe content similar to that reported here as a low-limit value corresponds with those associated with the finding of small amounts of VHOC emissions, while the emission rates become saturated when enrichment with Fe(III) is relatively minor (Keppler et al., 2000). Saturation at relatively low soil Fe concentrations was also reported by Huber et al. (2009)." (lines 640-643).

L478: I would replace “merges” with “combines”

Answer: Table 2 (originally Table 4) is now introduced in Sect. 3.1, to support the discussion of flux magnitude with the statistical analysis that is incorporated into the table. We have also removed the F:C parameter from the table (according to one of the following comments), such that we do not use "merges" in the revised sentence (line 408).

L481: “samplings” should be “sample”, and I would encourage revising this sentence to something like: “While the number of samples collected at each site was limited, Table 4 shows that the fluxes....”

Answer: The sentence has been revised accordingly (lines 645-647; see response to next comment).

L482: I would replace “In both” with “For the...sites,...”

Answer: The sentence has been revised according to this and the previous comment: "While the number of samples collected at each site was limited, Table 2 and Fig. 4 indicate elevated positive fluxes for the SD sites, and to some extent also at COAST-EGD-MD, with respect to both statistically significant and non-statistically significant positive fluxes" (lines 645-647).

L484: replace “in” with “at” before COAST-EGD-MD

Answer: Corrected (line 646; see answer to previous comment)

L485: A comma is needed after “winter”

Answer: Corrected (line 648)

L505: For consistency, replace VSLS with VHOC.

Answer: This text has been deleted, because it is less strongly supported when the statistical uncertainties in our analyses are taken into account. We generally use VHOCs through the text instead VSLS, except for the Abstract, Introduction and twice in the Results and discussions, where we specifically refer to VSLS (lines 347 and 358).

L506-07: Replace “during” with “at” before the site abbreviations.

Answer: We have separated the sentence into two sentences and revised them: "COAST-EGD-SD-s was associated with the highest incidence of both statistically significant and non-significant positive fluxes. Fluxes at COAST-EGD-SD-w were generally lower and with a smaller incidence of positive fluxes." (lines 665-667).

L515-19: Do you need the F:C ratio really aid in understanding these processes?

Answer: The F:C ratio has been removed from the table, because we are also able to support our discussion without it, and because it makes the table easier to extract information from (see also response to the next comment).

L544: Table 4. General comment: This is a hard table to read and extract information from - I almost feel as though presenting this graphically would be more impactful allowing the reader to see the trends rather than sifting through a lot of numbers that appear to vary greatly.

Because everything is bolded in the summary portion of Table 4, the rows should be explicitly labeled as to what the values are.

Answer: We have removed F:C from the table and we believe that this makes it easier to extract data. Information in the summary section of the table has been changed (including information which relates to statistical significance, instead of the F:C information), and we also explicitly label the rows in the summary portion. Note that Table 2 (originally Table 4) has been moved to Sect. 3.1 to support the discussion of fluxes from the various sites with the statistical analysis, which is incorporated into the table. The table now appears on line 494.

L553: Awkward as written, say something like: The results presented in Table 4 show that a higher... ”

Answer: Amended (lines 698-699)

L554: comma is needed after CHCl3

Answer: The sentence has been revised according to the updated analysis which takes into account the statistical uncertainties: "Differently than previous studies, brominated VHOCs had relatively higher overall incidence of positive fluxes than chlorinated VHOCs (Table 2)." (lines 698-699).

L555: “tends” should be “tended”

Answer: The sentence has been revised, also based on the updated analysis, which takes into account the statistical uncertainties of a site as a net source for the species (lines 699-702).

L558: I would suggest deleting the following (not needed): “suggesting both high emission and their balance to some extent by sinks for this species.”

Because there were watermelon fields, was there any harvesting or drying and decomposing plant material in the vicinity of the sampling? This can be a source of an array of halocarbons, particularly gases like CHCl3 and CHClBr2.

Answer: The whole sentence has been deleted, because it is less strongly supported by the updated statistical analysis. There was no harvesting or drying during the measurement periods. Overall positive net fluxes from the agricultural field site were not elevated compared to the bare soil sites, and tended to be, in most cases,

comparable to or lower than those from the natural vegetation (TMRX–ET) (see new Fig. 4).

L563: I would replace “are in general” with “were”

Answer: Amended (line 706)

L566-70: Please revise – it is unclear what you are trying to say.

Answer: We have revised the specified text for clarity: "The latter explanation may be supported by the fact that Albers et al. (2017) did not find any correlation between CHCl_3 emission rate and organic Cl in the soil. Furthermore, our study points to higher emission rates and incidence of VHOCs, and generally also of trihalomethanes, closer to the seawater (COAST–EGD and COAST–TKM sites), which suggests higher sensitivity to soil halide content than OM (Sect 3.2.2)." (lines 709-714).

L577-9: revise to something like: “...emission rates from both bare and vegetated soil sites supports the work by Albers et al. (2017) concerning the emission of trihalomethanes from the soil after trihaloacetyl hydrolysis (Table 3).”

Answer: Thank you. The text has been revised accordingly (lines 721-723). Note that the natural vegetation site is not statistically significantly a source for the trihalomethanes (Table 2).

L584: Agricultural emissions, such as from the watermelon farming, could be such a source. More details regarding the scale and influence of these operations would be useful.

Answer: While emissions of both CHBr_3 and CHBr_2Cl were observed at the agricultural field, even higher emission rates for these species were observed at the coastal sites, and for CHBr_2Cl also from bare soil (BARE–MSMR-2, BARE–MSD-2, BARE–MSD-3 and BARE–MSD-4; see Table 2 (originally Table 4) and new Fig. 4), where there is no agricultural activity. Based on this comment, we now include the following: "note that agriculture could potentially be a source for the emission of CHBr_2Cl and CHBr_3 for WM–KLY, but not for the other sites (Sect. 2.1.1)." (lines 729-731). Further, referring to the flux correlations between CHBrCl_2 , CHBr_2Cl and CHBr_3 , we mention in Sect. 3.3 that: "Note that these correlations can potentially be attributed to agricultural emission, considering that WM–KLY, but not TMRX–ET, was identified as a statistically significant source for the three trihalomethanes" (lines 848-850).

L589: replace “in” with “at” before the site name

Answer: Amended (line 737)

L589-91: Please revise the following – awkward as written: “No clearly more elevated positive flux of brominated compared to chlorinated trihalomethanes was observed for this site...”

Answer: The sentence has been rephrased (lines 737-740).

L600: include (Table 4) to direct the reader to this information

Answer: Done (line 748)

L601: For the statement “...indicating strong emission and deposition...”, if the flux is positive, then the emissions outweigh the deposition or other loss processes - revise to clarify your point. Figure 2 counters the point of “strong deposition” for the methyl halides.

Answer: The sentence has been revised based on this comment and the updated analysis that takes into account statistical uncertainties: "A relatively high incidence of negative fluxes was observed for CH₃Br, and more statistically significantly so for CH₃Cl and CH₃I, implying high rates of both emission and deposition, at least for the latter two, in the studied area (Table 2)." (lines 746-748).

L604-05: Cultivated watermelon fields (agricultural emissions) are different from local vegetation, please distinguish as such.

Answer: "local vegetation" has been replaced with "agricultural field" (line 753).

L644-46: please revise, reads awkwardly

Answer: The text has been revised: "It should be noted, however, that the fluxes that we used for the methyl halide emission efficiencies were based on measured net flux rather than measured emission flux. This might also explain the inconsistency between the relative CH₃I-emission efficiency calculated by Keppler et al. (2000) and by us" (lines 795-798).

L662-663: How are the data grouped for Table 5? Is this simply for all sampling heights lumped together? Is there a difference when grouped by height? Replace “evaluated” with “measured”

Answer: Table 4 (originally Table 5) refers to lumped correlations, to avoid higher correlations due to systematic trends of mixing ratios with height. Where height was taken into account, correlations tended to be higher and more statistically significant, but this does not well represent the correlations between species. Pursuant to this comment, we have added the following to the caption of Table 4: "Correlations were calculated for mean mixing ratios at each site" (line 832). The sentence has been revised: "Table 4 presents the Pearson correlation coefficients (r) between the measured mixing ratios of VHOCs at the Dead Sea, separately for all sites and for the terrestrial sites only, as well as separately for BARE, COAST, and the natural vegetation and agricultural field sites (VEG). For COAST, r is also presented individually for the two sites which were closest to the seawater (SD)." (lines 813-817).

L670: Please consider revising: “...reinforce predominant contribution of VHOCs from terrestrial sources...” – I would consider this to be an overstatement.

Answer: The sentence has been revised: "Correlations were in most cases either similar or smaller when we included measurements from the seawater site SEA-KDM, which may reinforce the notion that emission from the seawater does not contribute significantly to VHOC mixing ratios in the area of the Dead Sea." (lines 825-828).

L672: The r2 values are quite low, and without being able to see the correlation plots of these gases, it is difficult to adequately assess the commonality of their sources and sinks. How do these specific r2 values translate in to common sources and sinks?

Answer: In the revised version, we indicate the *p*-value associated with the presented correlation values. We also present the correlations individually for the different site

types. This enables us to better support the correlation between CHCl_3 and methyl halides, at least for some of the site types (Table 4). Because r values (we use Pearson correlation coefficient (r) rather than coefficient of determination (r^2) in the revised version) cannot be directly translated into common sources and sinks, the short discussion about correlations according to Table 4 is followed by a discussion based on the flux analysis (Table 2) and flux correlation analysis (Table 5). In particular for the correlation between CHCl_3 and the methyl halides, the related discussion is as follows: " Interestingly, in agreement with Table 4, Table 5 also shows relatively high correlations between CHCl_3 and all methyl halides, particularly for the BARE sites (CH_3I , $r = 0.68$, $p < 0.15$; CH_3Br , $r = 0.83$, $p < 0.05$; CH_3Cl , $r < 0.86$, $p < 0.05$), and SD sites (CH_3I , $r = 0.99$, $p < 0.05$; CH_3Br , $r = 0.59$, $p > 0.15$; CH_3Cl , $r = 0.91$, $p < 0.1$). Remarkably, a high correlation was found for CH_3I with CHCl_3 and C_2HCl_3 at the SD sites ($r = 0.99$, $p < 0.05$ in both cases). Positive fluxes of the three species were observed at the SD sites in most cases, although with only moderate statistical significance for CHCl_3 (Table 2). Weissflog et al. (2005) found that emission of C_2HCl_3 , CHCl_3 and other chlorinated VHOCs can occur from salt lakes via the activity of halobacteria in the presence of dissolved Fe (III) and crystallized NaCl. The strong correlations of CHCl_3 , C_2HCl_3 and CH_3I at the SD sites, where statistically significant fluxes were frequently measured for these species, reinforce the colocalized emission of CHCl_3 and C_2HCl_3 from salt lake sediments, as indicated by Weissflog et al. (2005), and suggest that CH_3I can be emitted in a similar fashion. The fact that the relative emission efficiency of CH_3I in our study was much higher than under the conditions used by Keppler et al. (2000) supports the possibility that mechanisms other than the abiotic emission pathway proposed by Keppler et al. (2000) influence the emission of CH_3I at the Dead Sea (Sect. 3.2.3)." (lines 883-898). Hence, we believe that the integrated analyses indicate a common source for CHCl_3 , C_2HCl_3 and CH_3I . Note that, considering this comment, we removed the original sentence from the manuscript.

L678: Replace "records" with something like "shows"

Answer: Amended (line 838)

L680-81: Change "For the two last, ..." to something like: "For the latter two sites, ..."

Answer: Amended (line 840)

L685: Replace "demonstrates" with something like "shows" or The results in Table 6 show/illustrate...

Answer: Amended (line 843)

L692-94: Can you please expand upon the correlations being attributable to "common sinks" – what are the sinks and how is this driving the correlations?

Answer: Taking into account statistical uncertainties in our revised analyses indicated that the cultivated and natural vegetation could not be classified as statistically significant as either sink or source for methyl halides. Therefore, considering also this comment and the length of the discussion in Sect. 3.3, we have removed this specific discussion.

L740: replace "common emission" with something like "co-located emissions"

Answer: Done (line 893)

L826-27: I would recommend revising or omitting the following: "...from saline soil and salt lakes in stratospheric and tropospheric chemistry,..." , as there were no linkages made to how the compounds measured for this work play in to the local/regional/global budgets of tropospheric or stratospheric Cl, Br or I.

Answer: We agree, and the text has been revised accordingly: "Overall, along with other studies, the findings presented here highlight the potentially important role of saline soil and salt lakes in VHOC emission, and call for further research on VHOC emission rates and controlling mechanisms, and implications on stratospheric and tropospheric chemistry." (lines 974-977).

References

- Albers, C. N., Jacobsen, O. S., Flores, E. M. M., and Johnsen, A. R.: Arctic and Subarctic Natural Soils Emit Chloroform and Brominated Analogues by Alkaline Hydrolysis of Trihaloacetyl Compounds, *Environ Sci Technol*, 51, 6131-6138, 10.1021/acs.est.7b00144, 2017.
- Butler, J. H., King, D. B., Lobert, J. M., Montzka, S. A., Yvon-Lewis, S. A., Hall, B. D., Warwick, N. J., Mondeel, D. J., Aydin, M., and Elkins, J. W.: Oceanic distributions and emissions of short-lived halocarbons, *Global Biogeochem Cy*, 21, Artn Gb102310.1029/2006gb002732, 2007.
- Grubbs, F. E., and Beck, G.: Extension of Sample Sizes and Percentage Points for Significance Tests of Outlying Observations, *Technometrics*, 14, 847-&, Doi 10.2307/1267134, 1972.
- Huber, S. G., Kotte, K., Scholer, H. F., and Williams, J.: Natural Abiotic Formation of Trihalomethanes in Soil: Results from Laboratory Studies and Field Samples, *Environ Sci Technol*, 43, 4934-4939, 10.1021/es8032605, 2009.
- 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.
- Quack, B., Atlas, E., Petrick, G., and Wallace, D. W. R.: Bromoform and dibromomethane above the Mauritanian upwelling: Atmospheric distributions and oceanic emissions, *J Geophys Res-Atmos*, 112, Artn D0931210.1029/2006jd007614, 2007.
- Shafir, H., and Alpert, P.: Regional and local climatic effects on the Dead-Sea evaporation, *Climatic Change*, 105, 455-468, 10.1007/s10584-010-9892-8, 2011.
- Sive, B. C., Varner, R. K., Mao, H., Blake, D. R., Wingenter, O. W., and Talbot, R.: A large terrestrial source of methyl iodide, *Geophys Res Lett*, 34, Artn L1780810.1029/2007gl030528, 2007.
- 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.

Zhou, Y., Varner, R. K., Russo, R. S., Wingenter, O. W., Haase, K. B., Talbot, R., and Sive, B. C.: Coastal water source of short-lived halocarbons in New England, *J Geophys Res-Atmos*, 110, Artn D2130210.1029/2004jd005603, 2005.

Zhou, Y., Mao, H. T., Russo, R. S., Blake, D. R., Wingenter, O. W., Haase, K. B., Ambrose, J., Varner, R. K., Talbot, R., and Sive, B. C.: Bromoform and dibromomethane measurements in the seacoast region of New Hampshire, 2002-2004, *J Geophys Res-Atmos*, 113, Artn D0830510.1029/2007jd009103, 2008.