Response to Referee #2

We would like to thank reviewer 2 for reviewing the manuscript. Below you find our point-by-point answers to your comments (*highlighted in italic*).

General Comments:

Referee #2:
The manuscript is reasonably well written but at times a little difficult to follow, owing in part to acronyms and repeated recitation of numbers, but also to text that seems to ramble without a clear focus in some places.

Author's response:
We have carefully revised the text taking your suggestions into account. Thus, we changed in particular Sections 3.2-3.5, 4 and 5, reduced the acronyms and repeated numbers, which are already listed in the tables. Further details can be found in our answers to your specific comments below.

The sampling approach appears sound, the data look to be good, and the approach is interesting. I'm wondering, however, if the authors are not letting the "trees get in the way of seeing the forest", in that in some places the detail is killing the main message.
Using a largely meteorological approach, the authors make the case that the ocean makes only a small contribution to the amount of gas in the boundary layer immediately above it. While that may be true for any of the 400 m2 boxes they use, I'm not so certain it is true for Peruvian upwelling altogether. At least, that has not been made clear to me. The point is made that most of the halocarbons over a given area are advected in, but it belies the possibility (probability?) that any elevation of concentration in a particular box coming from upwind portions of the upwelling zone contains air that has already been impacted by oceanic emissions. One could walk away from this paper thinking that the emissions of short lived halocarbons in upwelling regions like Peru are not that significant when, based on comparison with boundary layer burdens over the open ocean, it seems that they are. This is not to say that the analysis in this paper is not useful, but it does need to be put in perspective. The authors also need to discuss the tenuous nature of their assumption of steady state in a dynamic boundary layer and its implications to their conclusions.

We agree with the reviewer that the paper should avoid the impression, that the emissions of short lived halocarbons in upwelling regions like Peru are not that significant when, based on comparison with boundary layer burdens over the open ocean, it seems that they are. We also agree with the reviewer that the boundary layer burdens over the upwelling are much more significant than over the open ocean which is generally due to elevated oceanic concentrations and emissions in conjunction with stable layers in the atmosphere above upwelling regions. However during M91, the oceanic emissions, which were calculated along the cruise, are indeed generally not sufficient to explain the observed elevated atmospheric mixing ratios in the MABL, which therefore need advection. We add a more detailed description of the observed atmospheric phenomena and methodological constraints in the answer to the reviewer (below) and in the manuscript and therewith hope to better clarify our observations and interpretations of them.

Figure 3e of the manuscript shows that the observed atmospheric mixing ratios of all compounds show background concentrations of around 0.6 ppt for CH_3I, 1 ppt for CH_2Br_2 and 2 ppt for $CHBr_3$ along the whole cruise with varying elevations in the upwelling regions. The mixing ratios for CH_2Br_2 and $CHBr_3$ generally double (increase by 1 or 2 ppt above the direct Peruvian upwelling), while CH_3I is 3 to 4 times higher. The observed mean fluxes during the cruise can explain around 0.6 ppt for methyl iodide for an average MABL height of 300 m using a mean residence time of 7hr (derived from our FLEXPART trajectories), and 1.2 ppt under the average trade wind inversion of 1000 m using a mean residence time of 48 hrs. For the low fluxes of CH_2Br_2 and $CHBr_3$ the contribution to the MABL box is only between 0.1 and 0.3 ppt. Thus, while CH_3I-emissions can explain the local background, CH_2Br_2 and $CHBr_3$ fluxes cannot. Therefore, we believe that the bromocarbon background mixing ratios of 1 to 2 ppt in the MABL have to be advected, with the southerly low level flow parallel to the coast and below the trade inversion before the air masses reach the well-defined isolated MABL above the Peruvian upwelling. We also agree that a different emissions scenario before the cruise and in waters close to the cruise track may partly explain the discrepancy. However, we believe that during the time of the cruise a representative area of the Peruvian upwelling from 6 °S to 16 °S and 10 nm to 90 nm away from the coast was investigated. Surprisingly high sea surface concentrations and emissions in the upwelling waters different to those measured during the cruise appear unlikely to us. Next to this we even derive from observations that the ocean was mainly a sink for $CHBr_3$ between Dec 21 and 25. Also possible elevated emissions from coastal sources, not measured during the cruise, likely add to the atmospheric mixing ratios. Indeed elevated coastal emissions could

explain the threefold increase of $CHBr_3$ atmospheric mixing ratios on December 25 with SE wind from the coast (Figure 3), in contrast to Dec 17 when the ship passed the same coastal position but revealing lower VMR and a southern flow. Additionally, direct above the upwelling regions the MABL can be as low as 10 m and shows a mean of 100 m (Figure 4c). Thus local emissions mix into a much lower boundary. As an example an emission of 1000 pmol m^{-2} hr^{-1} explain an atmospheric mixing ratio increase of 1.75 ppt during a residence time of 7 hr and a MABL height of 100 m. These relatively high emissions have occasionally been observed during the cruise for methyl iodide and the resulting increase in mixing ratios can be even higher in lower MABLs. These high emissions have however barely been observed for $CHBr_3$ and CH_2Br_2 and are also unlikely due to the low water concentrations and low wind speeds observed during the cruise. Thus, we conclude that coastal contributions with highly elevated emissions of e.g. macro algae are very likely as additional sources for the bromocarbons.

Overall we conclude that the variable and elevated atmospheric mixing ratios observed above the East Pacific and the Peruvian upwelling region, including both open and coastal ocean, can be explained by a mix of oceanic sources and atmospheric phenomena.

In addition the mechanism and concept of the air-sea exchange contribution could be different in the coastal upwelling, e.g. the stable low MABL, which leads to suppressed vertical mixing of surface air, could lead to a gradient of the near surface VSLS with height. Thus, the surface VSLS observations may therefore only be representative for the low and not for the entire MABL column. Next to these considerations, the very low MABL events above the cold, near-coastal waters,which theoretically increase the oceanic contribution to the atmosphere could often not be taken into account in the source/loss estimates due to the low resolved topography in ERA-Interim affecting the trajectory modelling (see manuscript: Section 2.5, Figure 3d). Finally, we also agree that our steady state assumption for the individual box model calculations in the MABL could also be affected by dynamical meteorological fluctuations (e.g. wind and MABL changes). These fluctuations were however quite rare during the time of our ship cruise. Overall, we think that follow-up studies are needed for the East Pacific regime including more marine observations also resolving MABL gradients and high resolution transport modelling including the coast lines.

We add this discussion to Section 4 and an outlook in Section 5 to better address these issues and to put our Peruvian Upwelling analysis in a broader perspective.

Finally, while the authors do address the uncertainties of ocean concentrations and atmospheric mixing ratios, I'm concerned about the uncertainties introduced by the modeled components, e.g., advection, degradation, air-sea exchange, and how they might impact the authors' conclusions. These seem to be ignored. The authors should at least discuss this, if not address it quantitatively.

We agree and will include a discussion of the method uncertainties into the existing discussion in Section 4:

"Uncertainties may result from the applied method, which accounts for a 400 m^2 box around a measurement point assuming steady state. The cruise track covered a significantly large area of the Peruvian Upwelling between 5° S and 16° S and higher elevated seas surface concentrations and emissions are not to be expected during these rather stable meteorological conditions. Additional uncertainties in our source-loss estimate may arise from deficiencies in the meteorological input fields from ERA-Interim reanalysis as well as from the air mass transport simulated by FLEXPART. Both could lead to either a shorter or longer residence time of the surface air masses within the MABL or below the trade inversion and thus influence the COL term. In particular very close to the coast, where the source-loss estimate could not be applied due to the trajectory analysis gaps (Section 2.5), the ODRs of the compounds might be different. Here potential high coastal emissions in combination with stable atmospheric stratification leading to slow vertical transport into the free troposphere, could significantly increase the oceanic contribution to the MABL and to the atmosphere below the trade inversion and explain the elevated atmospheric mixing ratios. In addition, different parameterizations for the wind-based transfer coefficient k_w, as discussed in Lennartz et al. (2015) and Fuhlbrügge et al (2015) in more detail, can impact the air-sea gas exchange and thus the ODRs. Applying the k_w parameterizations of Liss and Merlivat (1986) as well as Wanninkhof and McGillis (1999) lead both to mean ODRs in the MABL of 0.02 (bromoform), 0.07 (dibromomethane) and 0.21 (methyl iodide) and below the trade inversion of 0.08 / 0.08 (bromoform), 0.25 / 0.26 (dibromomethane) and 0.69 / 0.75 (methyl iodide) and thus an even lower oceanic contribution to the atmosphere in this region. Further uncertainties may arise from variations of the MABL VSLS lifetimes and thus the chemical degradation of the compounds we use in this study. This would affect the computed advection (ADR) and not the oceanic contribution."

Specific Comments:

1. The repeated use of similar acronyms requires that the reader keep looking back at the text. This could be helped considerably with a labeled diagram of the boundary layer box and its fluxes over the 400 m2 ocean surface.

We agree and add this diagram of the processes into the manuscript:

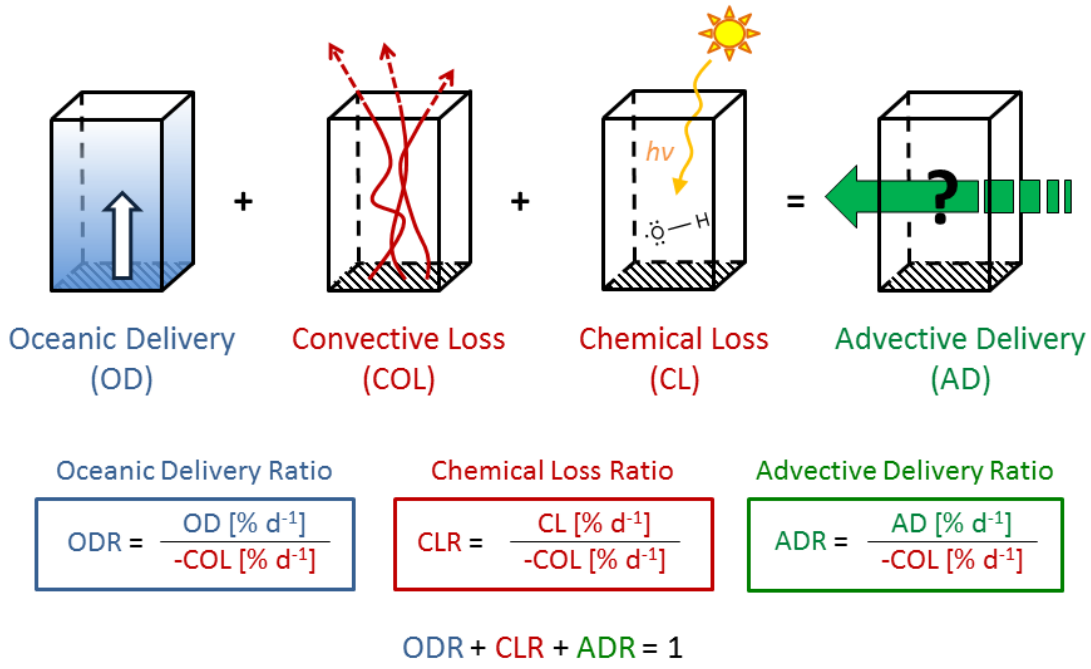

Figure 1: Schematic summery of the components of the applied mass-balance concept: Oceanic Delivery (OD), the Convective Loss (COL), the Chemical Loss (CL), the Advective Delivery (AD), the Oceanic Delivery Ratio (ODR), the Chemical Loss Ratio (CLR) and the Advective Delivery Ratio (ADR). The shaded area reflects an area of 400 m^2.

2. P. 20599, line 1, replace "more" with "other"

Done.

3. P. 20600, line 13-14, delete "respectively"

Done.

4. P. 20600, lines 15-19, delete all

Since this part gives an overview of the manuscript structure we would like to keep this part.

5. P. 20600, line 23, replace "ships" with "ship"

Done.

6. P. 20601, line 21, replace "from" with "near"

Done.

7. P. 20603, line 3, how high is the 5th superstructure, what obstacles surround it, etc.?

The 5th superstructure deck on R/V METEOR is found at about 20m above sea level. A smaller 6th superstructure deck with additional masts is found above it. More information can be found at: https://www.ldf.uni-hamburg.de/en/meteor/technisches.html.
A jib was used to sample the VSLS freely about 5 – 6 m portside, to avoid any ship influences on the measurements. We added this to the manuscript.

8. P. 20603, line 11, define "moon pool"

A moon pool (hydrographic shaft) is an opening in the base of the ship hull that gives access to the water below, providing protection for the sampling instruments. We added this information to the manuscript:
"102 water samples were taken 3 hourly at a depth of 6.8 m from a continuously working water pump in the hydrographic shaft, an opening in the base of the ship hull of R/V Meteor, after December 9, 2012."

9. P. 20604, lines 10,11, remove sentence, place refs at end of previous sentence.

Done.

10. P. 20604 lines 22, 23, move "from the ocean surface to right after "launched"; make "positions" singular.

Done.

11. P. 20606, line 1, replace "relating" with "ratioing"; replace "receive" with "estimate".

Done.

12. P. 20606, line 3, delete "respectively"

Done.

13. P. 20606, lines 1-5. This part is confusing and a good place where the diagram could be put to use.

We will insert the new diagram at this point.

14. P. 20606, line 23, how much of a temperature drop was it to get to 18 degrees?

This depends on the region. Generally outside the upwelling area, the SST ranges between 19 and 22 °C. We added this information to the manuscript:
"The most intense upwelling is observed for several times near the coast where both, SST and SAT rapidly drop from 19 – 22 °C to less than 18 °C (**Fehler! Verweisquelle konnte nicht gefunden werden.**a)."

15. P. 20607, lines 1-10, Could this be reduced to a simple sentence?

We think that the relation between absolute and relative humidity and their indication of different air masses might not be trivial for all readers and thus like to keep this part in the manuscript.

16. P. 20607, lines 12-18, Is this paragraph necessary? Similarly, is the next one necessary? Can the authors simply make a few statements about the meaning of these concentrations, how they relate to other areas, and how they might be useful to support their conclusions? Numbers are best placed in tables so the authors can refer to them and keep the text focused on the issues at hand.

We agree with the reviewer and shortened the first and second paragraph accordingly. We also cut out repeated numbers and refer to the responding tables.

"Surface bromoform concentrations in the Peruvian upwelling are generally lower during the cruise compared to the Mauritanian upwelling while dibromomethane surface water concentrations are comparable. However, methyl iodide concentrations are almost 8 times higher than in the Mauritanian upwelling (Figure 3d, Table 1, Hepach et al., 2014). Samples taken in the upwelling areas show elevated concentrations compared to the open ocean for all compounds. For further discussion on the distribution of the oceanic halocarbons, see Hepach et al. (2016, submitted to ACPD).

Atmospheric mixing ratios of $CHBr_3$ are on average 2.91 ± 0.68 ppt (Table 1). CH_2Br_2 mixing ratios of 1.25 ± 0.26 ppt are low and show a similar temporal pattern with bromoform (Table 3). Mixing ratios of both compounds are significantly lower above the Peruvian upwelling compared to observations above the Mauritanian upwelling, while CH_3I mixing ratios are comparable (Fuhlbrügge et al., 2013). Elevated mixing ratios for all three compounds are generally found above intense cold oceanic upwelling regions close to the Peruvian coast (Figure 3e). While the bromocarbons double above the upwelling, methyl iodide mixing ratios increase up to 5-fold, showing its stronger accumulation in the low boundary layer.

The concentration ratio of dibromomethane and bromoform can be used as an indicator of fresh bromocarbon sources along coastal areas. Low ratios of about 0.1 have been observed at coastal source regions and are interpreted as the emission ratios of macro algae (Yokouchi et al., 2005;Carpenter et al., 2003). The applied shorter mean lifetime of bromoform (15 days) in contrast to dibromomethane (94 days) in the boundary layer after Carpenter et al. (2014)

leads to an increase of the ratio during transport as long as the air mass is not enriched with fresh bromoform. A general decrease of the concentration ratio is found from the North to the South during the cruise (Figure 3f), implying relatively remote air masses in the North and an intensification of fresh sources towards the South, which is also reflected by elevated water concentrations towards the South. Atmospheric methyl iodide measurements along the cruise track reveal a mean mixing ratio of 1.54 ± 0.49 ppt, which, similar to the two bromocarbons, maximizes over the coastal upwelling regions (Figure 3e)."

17. P. 20608, lines 23-25, This is a strange statement in that it is obvious. MBL concentrations are always a function of air-sea differences, in situ loss, and advection. The sentence after this is equally unnecessary.

We agree and removed the sentences.

18. P. 20609, all, This tutorial discussion could be condensed to a few sentences, in my mind. If the authors are trying to explain the 15-16 December anomaly, they should focus on that, not ramble through all the rest.

In this part we describe the overall atmospheric conditions (not only 15-16 December), which significantly influence surface VSLS abundances in this region, and the methodology to explain how the missing MABL heights were calculated. Although we think this part is necessary we shortened the section according to the reviewer's suggestion and rewrote this section to:

"3.3 Lower atmospheric conditions

The relative humidity shows a strong vertical gradient from over 75 % to less than 50 % at ~1 km height (Figure 4a) which indicates an increase of the atmospheric stability with height due to suppressed mixing. This convective barrier, known as the trade inversion (Riehl, 1954, 1979;Höflich, 1972), is also reflected in the meridional wind (Figure 4b). Below ~1 km altitude the Southeast trade winds create a strong positive meridional wind component, also visible in the forward trajectories (Figure 2c-d). The flow of air masses in the Hadley Cell back to the subtropics causes a predominantly northerly wind above ~1 km height. The intense increase of θ_v in combination with the relative humidity decrease and the wind shear

at ~1 km height identifies this level as a strong vertical transport barrier (Figure 4c). Above the cold upwelling water, temperature inversions create additional stable layers above the surface, leading to very low MABL heights of < 100 m, e.g., on December 03, 08 or 17, 2012 and to a reduced vertical transport of surface air. The mean MABL height from the radiosonde observations is 370 ± 170 m (ERA-Interim 376 ± 169 m). The relative humidity, SAT, SST and wind speed are good indicators for the MABL conditions in this oceanic region and these meteorological parameters show significant correlations with the observed MABL height (Table 3). Thus, we use a multiple linear regression based on these parameters to estimate the MABL height above the coastal upwelling (Section 2.2.2). The regressed MABL heights above the cold upwelling regions are 158 ± 79 m and go down to even 10 m as was previously observed above the Mauritanian Upwelling (Fuhlbrügge et al., 2013). With the regressed MABL heights, the mean MABL height during the cruise decreases to 307 ± 177 m. The stable atmospheric conditions from the surface to the trade inversion lead to strong transport barriers and to a supressed transport of surface and MABL air into the free troposphere (Figure 4d)."

19. P. 20610, lines 9-16, This is good and relevant.

Thank you.

20. P. 20610, lines 25 ff, This is where the authors need to insert some perspective as discussed under General Comments. Also, how much of the discussion on the following page is relevant to their main point?

We included the following sentences on page 20611, line 2:
"Generally, the low ODRs along the cruise track are caused by the relatively low oceanic emissions. Since the observed atmospheric concentrations cannot be explained by the local oceanic emissions advection leads to the background concentrations of CH_2Br_2 and $CHBr_3$. According to the backward trajectories, potential source regions may be found closer to the coast and to the South. The elevations of the atmospheric mixing ratios above the cold coastal upwelling can partly be explained by accumulation of local oceanic emissions in the stable low MABL. However, as the emissions appear generally not strong enough, except for

methyl iodide, to explain the mixing ratios, the contribution of coastal sources is very likely (Figure 2b)."

And on page 20611, line 13:
"These numbers imply that observed VSLS concentrations are advected below the trade inversion in the more open ocean regions during the cruise. An overall discussion is given in Section 4."
The remaining sentences of section 3.4 are removed (line 13-20).

A more detailed discussion of the phenomena is given within the general comments to the Reviewer 2 above and in the detailed discussion in Section 4 of the manuscript.

21. P. 20611-12, Section 3.5. Numbers are getting in the way of the points the authors need to make. There is a table of correlation coefficients. The authors should make the important points and simply refer to the table.

We agree and removed the numbers from the text referring to the Table 3, thus the text in the manuscript reduces to:
"Fuhlbrügge et al. (2013) identified the influence of meteorological conditions, in particular of MABL height variations on VSLS abundances in the tropical Northeast Atlantic above the Mauritanian Upwelling, and suggested a general correlation of MABL conditions and VSLS abundances over oceanic upwelling regions. Indeed, we also find significant high correlations between meteorological parameters and the abundances of bromoform, dibromomethane and methyl iodide (Table 3) along the Peruvian coast. The predominantly moderate winds during the cruise are negatively correlated with the atmospheric VSLS and positively correlated with the MABL height. This shows that VSLS abundances tend to be elevated during periods of lower wind speeds which occur also lead to reduced mixing of surface air and therefore to lower MABL heights, in particular above the coastal upwelling events on December 11, 15-17 and 24, 2012, where local sources could accumulate even more. SAT and SST both are negatively correlated with atmospheric VSLS, since elevated atmospheric VSLS mixing ratios are generally found close to the oceanic upwelling regions with low SATs and SSTs. In these regions the decrease of the SATs leads to an increase of the relative humidity (section 3.1), resulting in a significantly high correlation between the surface relative humidity and the VSLS. Since SAT and SST impact the MABL, which affects the relative humidity, these

correlation coefficients are co-correlated with each other. Correlation coefficients between the MABL height and the VSLS are slightly lower (Table 3). A principle component analysis of the parameters in Table 3 furthermore underlined a strong connection between SAT, SST, MABL height, relative humidity and atmospheric mixing ratios of $CHBr_3$ and CH_2Br_2 (not shown here).

The results reveal that the MABL properties (height and stability) during M91 influence the VSLS abundances at the marine surface, although not as distinct as above the Mauritanian Upwelling during the DRIVE campaign (Fuhlbrügge et al., 2013). A comparison between the observations from this campaign and DRIVE (Figure 5) shows that the lower variance of observations during M91 may explain the lower correlation. Generally higher emissions and occasional lower and even more stable MABL-heights during DRIVE can explain up to 100% of the atmospheric abundances (Fuhlbrügge et al., 2013;Hepach et al., 2014), while during M91 the observed elevations could only partly be explained by the local oceanic emissions."

22. P. 20612-20614, Section 4.0, This is where an overall perspective of upwelling contributions and an understanding of overall uncertainties would really help. The authors make some good points here; I'd just like to see them better substantiated.

We agree and will therefore - in addition to the above discussed method uncertainties (reviewer comment 2) - include the following discussion on page 20614, line 5:

"The contribution of oceanic emissions to the atmospheric mixing ratios in the MABL of the Peruvian upwelling reveals to be rather low in its more open ocean area under the given meteorological conditions. While the cruise track covered a representative area of the Peruvian upwelling elevated oceanic VSLS emissions that could explain the generally high atmospheric VSLS were only observed for methyl iodide. Bromocarbon emissions would have to be two magnitudes larger to explain the observed VMR in the more open ocean regions and a magnitude larger in the direct coastal upwelling regions with low MABL heights. These observations of the brominated compounds need to include upwind advection of elevated sources from the South, and higher elevated coastal emissions not measured during the cruise, while dynamical fluctuations in emissions scenarios close to the cruise time and place may also have to be considered."

23. P. 20614-20616, too many numbers here. This should be a summary of the main points the authors are trying to make with their data, where the gaps in our understanding still are, what should be done to remedy those gaps, and why. The summary should be much shorter.

We agree and shortened the summary accordingly by i.e. removing most numbers and concentrating on the main points:

"This study investigated the contribution of oceanic emissions to VSLS abundances in the lowermost atmosphere as well as meteorological constrains on this contribution above both, coastal upwelling and open ocean along the Peruvian coast during December 2012. Meteorological data were obtained on R/V METEOR near the ocean surface and by radiosondes up to the stratosphere. Oceanic VSLS emissions along the cruise track were determined from air and surface water data. The transport of air masses was determined with FLEXPART trajectories.

Oceanic upwelling was observed close to the Peruvian coast. On average a low, stable MABL height of 307 ± 177 m was encountered during the cruise, decreasing to on average 100 m above the upwelling. A distinct trade inversion at 1.1 ± 0.3 km height evolved as the dominant transport barrier for MABL air into the free troposphere during the cruise. The halogenated VSLS bromoform and dibromomethane showed low oceanic emissions of 117 ± 492 pmol m^{-2} hr^{-1} for bromoform and 245 ± 299 pmol m^{-2} hr^{-1} for dibromomethane, while methyl iodide emissions were elevated with 856 ± 623 pmol m^{-2} hr^{-1}. The atmospheric mixing ratios of the compounds were elevated with 2.9 ± 0.7 ppt (bromoform), 1.3 ± 0.3 ppt (dibromomethane) and 1.5 ± 0.5 ppt (methyl iodide). The oceanic emissions along the cruise track explained on average only 3 % (-8 to 33 %) of bromoform, 10 % (-5 to 45 %) of dibromomethane, and 28 % (3 to 80 %) of methyl iodide abundances in the MABL. Thus, the expected significant contribution of local oceanic VSLS emissions from the Peruvian upwelling to the overlying atmosphere was not captured during the time and location of the cruise. The elevated atmospheric VSLS mixing ratios above the Peruvian upwelling therefore appear largely advected and enriched along the Peruvian coast before reaching the ship. Additional potential source regions must exist closer to the coast and also further South of the cruise track along the coast line. Nevertheless, significant correlations between the MABL height and marine atmospheric abundances of the VSLS reveal an impact of the oceanic emissions on the atmospheric VSLS mixing ratio variations.

Our study confirms that MABL height and stability are generally related with atmospheric VSLS abundances above oceanic upwelling regions. Additionally, a widespread trade

inversion can lead to a near-surface accumulation of the VSLS and thus also impact oceanic emissions. Despite the observed elevated atmospheric concentrations during the cruise, a significant contribution of oceanic emissions to the atmosphere, in particular of the bromocarbons $CHBr_3$ and CH_2Br_2, was not identified in the observed area during the time of the cruise. Further studies are necessary to clearly uncover the source regions of the elevated atmospheric VSLS in the Peruvian upwelling. Also the double transport barrier phenomena should be investigated in future studies of other oceanic upwelling regions as well. “

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