

General Comment to the Editor and Reviewers

We would like to thank the Reviewers for their thorough comments and questions that helped very much to improve the manuscript and to clarify some issues. In the following text, the full comments of the Reviewers are listed together with our responses (written in bold). Text, which will enter the revised version of our manuscript, is written in blue letters.

We inform the editor and reviewers that we had to correct the measurement uncertainty for the organic bromine (Br_{org}) from VLS. The values changed from ± 0.60 ppt to ± 0.44 ppt for the upper troposphere and from ± 0.60 ppt to ± 0.29 ppt at the LZRH. Therefore, the measurement uncertainty for the total organic bromine also changes. The value in the upper troposphere changed from ± 1.78 to ± 1.62 and from ± 1.78 to ± 1.47 at the LZRH. Table 10 and Table 12 have been updated with these values, as well as the corresponding passages in the manuscript.

Reply to anonymous Referee #1

Page 4958, line 4: . . . were collected . . . Same in the conclusions. Please check the use of grammar throughout the text. One can clearly tell that section 2.2.2 was probably written by a native English speaker compared to the rest of the text. Since you have 2 native English speakers in your co-author team I strongly recommend to let them correct the rest of the text, for better readability and to avoid some odd grammar. So please make use of your co-authors.

The paper has been re-edited by a native speaker to correct for grammatical errors and in order to ensure a better readability. This has been done in a way not to change any statements or discussion presented in the original manuscript.

Page 4958, line 23: Who do you mean by our group, since actually 3 groups are listed in the affiliations?

In this context, “our group” means the group from Goethe University Frankfurt (GUF). Changed in the text.

P 4958, l 26: . . . factor of about 60. . . : Is this statement true throughout the stratosphere, or which part does it refer to?

The factor calculated by Sinnhuber et al. (2009) is 64 for the global annual mean. The model calculations for this so called alpha – factor cover an altitude from 10-50 km (see e.g. Fig. 8 in Sinnhuber et al. (2009)).

“On average bromine atoms are more effective by a factor of 64 than chlorine atoms in destroying ozone in the altitude range from 10-50 km (Sinnhuber et al., 2009)”

P 4959, I 19: . . .well correlated. . . : What do you understand under well correlated? And are you talking about a linear correlation?

In this respect well correlated means that there is an obvious relationship between two compounds. The r^2 values given by Brinckmann et al. (2011) for the correlations of CHBr_3 with CH_2Br_2 , CHBr_2Cl and CHBrCl_2 are 0.91, 0.97 and 0.87 respectively, whereas the r^2 value for the correlation of CHBr_3 with CH_2BrCl is only 0.42.

“With the exception of CH_2BrCl the mixing ratios of all of these bromine carrying compounds show linear correlations in the troposphere with Pearson product-moment correlation coefficients (R) mostly greater than 0.8 (e.g. Brinckmann et al., 2012).”

P 4959, I 25: . . . in the atmosphere: Do you mean free troposphere, or UT here? I guess there are observations of PGs in the boundary layer and in the stratosphere. Or which PGs do you mean? Only organic?

This statement is true for both organic and inorganic PG in the upper troposphere and TTL.

“The fate of the organic and inorganic product gases from the loss reactions in the upper tropical troposphere and TTL is still largely unknown as so far no observations of these are available in this region of the atmosphere. To our knowledge there is only a single balloon profile showing BrO close to the detection limit (Dorf et al.,2006; Montzka and Reimann et al.,2011).

P 4960, I 10: . . . are decreasing in the atmosphere. Please add a reference here.
Reference was added.

“For the future, an increasing relative contribution to stratospheric chlorine and bromine from VSLs is expected as anthropogenic long-lived bromine (Newland et al., 2013) and chlorine (e.g. Montzka and Reimann, 2011) source gases are decreasing in the atmosphere.”

P 4961, I 5: . . .in Northern Borneo. Please add longitude and latitude here.

“Measurements flights were conducted from Miri in Northern Borneo and probing a wide geographical range (1°N - 8°N, 102°E - 122°E), different geophysical conditions and altitudes from the planetary boundary layer up to 13 km.”

P 4962, I 5: rationales? What do you mean? Seems an odd wording to me. See also comments on Table 1 below.

Changed “rationales” to “mission objectives”.

P 4962, I 6: 500? In the abstract you state 700. Or do the 700 refer to the total of both instruments?

The 500 is for the GHOST-MS instrument, the 700 is for GHOST-MS and WASP together.

P 4962, I 17-19: Not needed. Has been stated and explained before. I suggest to remove the entire sentence.

We agree with your comment, sentence is removed.

P 4967, I 15: Since you quantified the memory effect: Can you give explicit numbers here.

The correction is different for every data point as it depends on the change in mixing ratio between two measurements. Typical numbers are given in the paragraph further below.

“The quantification is based on the difference in concentration to the previous measurement. The entire procedure for the corrections is described in detail in Sala (2014).

P 4968, I 29: What do you mean by boundary layer altitude? The boundary layer height/thickness? Or the altitude of the top of the boundary layer?

In this context we mean altitude of the top of the boundary layer.

“The radiosonde launches give information about the top of the boundary layer, which is somewhat away in space and time from the different flight tracks.”

P 4970, I 17: Can you quantify your reasonable agreement?

” For the mixed bromochlorocarbons overall reasonable agreement is observed. The differences between the two datasets are lower than 15% for all substances and within their measurement uncertainties.”

P 4973, I 12-15: How do your higher values of CH₂BrCl compare to the WASP samples for that particular flight?

“These exceptional values are a factor of 2.5 higher than the mixing ratios measured during the rest of the campaign, but the simultaneous samples of the WASP instrument do not corroborated these GHOST-MS observations. Therefore, it cannot be excluded, that this was a measurement error of the GHOST-MS instrument, even though we have no indication for a malfunction during that particular flight.”

P 4975, I 1-3: Please check the grammar – not sure what you mean here.

“The mixing ratio in the UT is the second lowest of all species discussed here. Regarding the effect of CHBr₂Cl on stratospheric ozone depletion, this substance has nearly the same impact as the sum of the two longer lived minor VLS CHBrCl₂ and CH₂BrCl. This is due to the fact that CHBr₂Cl carries two bromine atoms while the other two minor VLS carry only one.”

P 4976, I 27: Which sources? Do you have any suggestions/speculations?

We cannot determine which specific source was relevant from our measurements, but in general the sources of CHBr₃ are mainly from coastal regions and oceanic upwelling regions (e.g., Quack and Wallace, 2004; Brinckmann et al., 2012; Leedham et al., 2013).

P 4981, Paragraph 3.6: Please also list the total Br_{org} values here. From Table 10 on page 5001 I also calculate different % values for the VSLs contribution – 19% for WMO and 22% for SHIVA. Please check the number and correct accordingly.

The total Br_{org} values have been added. Furthermore, we added one more digit to the percentage values given in Table 10. As a matter of course the budget has to be 100% in total, however some small rounding errors can occur.

P 4983, second and third paragraph: Please state all VSLs values from Table 12 also in the text at the appropriate place.

Values have been added to the text where appropriate.

P 4985, l 8: Add reference at the end of the sentence for the balloon-borne measurements.

Laube et al., 2008 and Brinckmann et al., 2012 have been added as references.

P 4992, Table 1: This post activity report mentioned in the header is not publicly available and also not listed in your list of references. What do you mean by Rationale? I guess the 4th column (Region) does not help any reader outside the SHIVA community. I suggest to either remove this column or to give some longitude, latitude range? Or you add a map where you indicate all these places.

We decided to leave out the citation of the post activity report and acknowledge the authors for providing the table instead. A flight map with the corresponding places was added as Figure 1. This was also suggested by the 2nd Referee.

P 5003, Table 12: Add an asterisk to your value for the Teresina 2005 observations published by Laube et al. and explain as stated in the text (Page 4983, line 25..) why these values probably deviate.

Added a further explanation: “Low VSLs values (*) found in Teresina (2005) are most probably due to sample loss in the canisters caused by the long period which elapsed between sampling and analysis of the air samples. “

Technical comments:

Page 4957: Affiliations 2 and 3: Why is Centre for Oceanic and Atmospheric Science listed twice?

Took out “Centre for Oceanic and Atmospheric Science” in Affiliation 2.

P 4959, line 5: . . .source gases (SGs). . . Also PGs further down. You use SGs sometimes and write source gases at other places. Once introduced you should use SGs in the following text.

Changed where appropriate.

P 4959, l 8: Change order of Brinckmann and Montzka, according to year.

Changed in the text.

P 4959, l 8: Sometimes Montzka et al. 2011 is used and at other occasions Montzka and Reimann et al., 2011 – please unify throughout the text.

Changed where appropriate.

P 4960, line 13: Eastern and Western Pacific – please check throughout the text: east, eastern, west, western . . . seem to be used arbitrarily
Changed where appropriate.

P 4963, l 26: . . . a novel . . . remove the 'a'.
Changed in the text.

P 4966, l 9 and l 15: GCMS or GC/MS? Please check throughout the text.
Changed where appropriate.

P 4967, l 1: remove: . . .in Miri.
Changed in the text.

P 4968, l 9 – l 10: Remove one of the two . . . marine boundary layer. . .
Changed in the text.

P 4969, l 23/24: Remove the sentence: Both instruments . . .
Changed in the text.

P 4970, l 4: Either introduce FT before (e.g. page 4968, line 7) and use throughout the text, or remove this abbreviation from the whole text
Changed where appropriate.

P 4977, line 4: Capitalize Western, like in the rest of the text
Changed in the text.

P 4979, l 21: Replace substances with VSLS
Changed in the text.

P 4985, line 16: Western Pacific
Changed in the text.

P 4998, Table 7, line 1: Add the year for WMO, SHIVA, CARIBIC, like in the other tables.
The year has been added.

All Tables and Figures: Not sure about ACP policy, but usually all labels and column headings should start with a capital letter, e.g. Time, Altitude, Mixing Ratio, Substance. .

We asked ACP editorial office and received the following answer: “The first word of a heading usually starts with a capital letter. The second word as in "Mixing ratio" is not capitalized. Our copy-editing will check that after the paper has been typesetted.”

Reply to anonymous Referee #2

General comments:

This paper presents new airborne measurements from organic bromine sampled above the Malaysian coast and the South China Sea during the SHIVA campaign 2011; published within the SHIVA ACPD special issue. This paper contains relevant new data which are suitable for the scope of ACP. The scientific results are presented in a clear and structured way; the use of English is appropriate. However, I am a bit disappointed by the lack of considering relevant new work, by the context of the background/ introduction as well as by the discussion and conclusions of the paper. Overall it became a bit boring data presentation, although an exciting unique data set for the Malaysian part of the tropical West Pacific exists! Here, the authors could have worked more thoroughly, searching through the new available publications (since WMO, 2011) as well as cross-linking their own work with other publication within the SHIVA consortium/ special issue also to get more inspired. A list of critical details is given below.

Due the amount and kind of corrections the paper may be publishable after a careful revision.

General comment on the review #2

The reviewer makes some very valuable comments which we discuss in detail below. The most serious general comment of the reviewer is a failure to include other relevant data for mixing ratios of VLS and their emissions into the Marine boundary layer (MBL). However, the MBL is not the focus of this paper and in particular the emissions are not, as our observation do not allow us to derive these. Our focus is on the observations in the free and upper troposphere and the transport into the stratosphere. Linking particular observations from the boundary layer with particular observations from the free troposphere is very difficult due to the difficulties in modelling the exact location, timing and magnitude of the convective transport. A paper trying to do this using our observations (Hamer et al., 2013) has just been rejected for publication in ACP, largely due to this difficulty. This is the reason why we do not want to study particular events but rather focus on typical values and budgets of total VLS bromine in the different altitude region of the western pacific area in which our observations were conducted. However, there are very few observations of the complete suite of the 5 VLS bromocarbons that we discuss here. The only other such studies known to us are the papers by Brinckmann et al. (2012) and Wisher et al. (2013) as well as the data compiled in the WMO report (Montzka and Reimann, 2011) which we discuss intensively in the paper. As absolute calibrations for most short lived bromocarbons have large uncertainties (see e.g. Butler et al., 2010), we also want to refrain from using observations which cannot be linked to the same calibration scales or which have not been intercompared.

We have added the Brinckmann et al. study in the table for the MBL values. We thank the reviewer for pointing this out. We have also made more reference to more recent work where adequate, as detailed below, however as very few studies focused on budgets and typical values, many of these discussion remain qualitative. In this we have largely followed the suggestions by the reviewer. However, we do not want to extend this study to discuss emissions and sources in detail.

Critical details:

The title should be changed as the SHIVA aircraft campaign did not cover the whole “tropical” Western Pacific but only Malaysian/South China Sea coastal and open waters!

We changed “Western Pacific” to “Western Pacific area” in the title to emphasize that not the whole Western Pacific is in the scope of this manuscript.

To be able to state that the “tropical” Western Pacific was investigated the authors would have to add more available and published data for that area including the HIPPO aircraft campaign (e.g. Tegtmeier et al., 2013 ACP SHIVA special issue), NOAA/ESRL ground station measurements (e.g. Hossaini et al., 2013 ACP SHIVA special issue) as well as the TransBrom ship campaign (Brinckmann et al., 2012; Tegtmeier et al., 2012).

At this time, no typical mixing ratios of brominated VSLs from the HIPPO campaign are published for the boundary layer. The paper of Tegtmeier et al. (2013) focus on methyl iodide. Hossaini et al. (2013) do not calculate a bromine budget or mean values for the boundary layer, free and upper troposphere. It is beyond the scope of the paper to present a summary of all available VSLs data which was obtained in that region in this manuscript (see also general comment on this review).

In principle the authors tend to often exclusively cite Montzka et al (2011) neglecting the original as well as newer publications since WMO (2011). They do, however, cite their own new papers Brinckmann et al. 2012 and Wisher et al. 2013. But then they neglect to cite their own open ocean tropical West Pacific VSLs measurements from Brinckmann et al (2012). Here a more thorough literature search and critical data comparison are needed, see the detailed comments below.

We checked the manuscript for more suitable and recent references then Montzka et al. 2011 and changed where appropriate.

I strongly suggest adding a map for the flight tracks to get an idea how spread out the SHIVA FALCON flights really were. This should be then Figure 1.

We agree with your suggestion and added a flight map, also to make clear where the Regions given in Table 1 are located, as this was also mentioned by the Referee #1..

Abstract:

P. 4958, lines 15-16: Rewrite the sentence “...could be a major course of brominated...”! If you want to refer to the “source” of VSLs you need to cite the production of VSLs in the ocean! I guess you mean here elevated atmospheric abundances around NE Borneo published by Pyle et al (2011).

“In contrast to the suggestion that the Western Pacific could be a region of strongly increased atmospheric VSLs abundance (Pyle et al., 2011), we found only in the upper troposphere a slightly enhanced amount of total organic bromine from VSLs relative to the levels reported in Montzka and Reimann et al. (2011) for other tropical regions..”

P. 4958, Line 22: “while being slightly higher” is a contradiction to the statement in line 15 “major source” > rewrite!

We agree with the reviewer that the formulation was misleading.

”From the SHIVA observations in the upper troposphere a budget for total organic bromine, including four halons (H-1301, H-1211, H-1202, H-2402), CH₃Br and the VSLS, is derived for the level of zero radiative heating (LZRH), the input region for the tropical tropopause layer (TTL) and thus also for the stratosphere. ”

Introduction:

I miss the background for the different oceanic sources of the different brominated VSLS, see comment below. Add this to the intro.

It is beyond the scope of the paper to distinguish between the different oceanic sources for the different brominated VSLS. Nevertheless, we added the following text to the introduction:

“The major bromine VSLS are dibromomethane (CH₂Br₂) and bromoform (CHBr₃), both having mainly oceanic sources like macroalgae (e.g. Baker et al., 2001; Carpenter et al. 2000; Carpenter et al., 2003),....” and “A detailed overview of the emissions of brominated halocarbons by tropical macroalgae is given in Leedham et al. (2013).”

P. 4959, lines 6-13: Rewrite this passage. I disagree with the message here that SG observations “...of VSLS have so far not been able to explain the amount of bromine derived in the stratosphere...” The three refs given here are not representing the current knowledge and substantial other references are missing for the VSLS part (line 8) e.g. Hossaini et al 2010; Liang et al 2010; Aschmann et al 2009; Tegtmeier et al 2012; Ziska et al. 2013; Hossaini et al. 2013...

With this statement we refer to the observations in the TTL, not throughout the troposphere. To make this clear, the passage was rewritten as follows:

“However, source gas observations of long-lived bromine compounds (Newland et al., 2013; Fraser et al., 1999) and VSLS in the tropical tropopause region (Laube et al., 2008; Montzka and Reimann et al., 2011; Brinckmann et al., 2012) have so far not been able to explain the amount of bromine derived in the stratosphere from observations of stratospheric BrO (e.g. Dorf et al., 2006; Montzka and Reimann et al., 2011).”

P. 4959, lines 10-13: Rewrite the whole sentence; this is not reflecting the current state of science here. We do have more insights of spatial and temporal variability of VSLS given the extensive fieldwork by ship and coastal measurements and observations based climatologies by e.g. Yokouchi et al; Carpenter et al., Pyle et al, 2011; Fuhlbrügge et al 2013 ACP SHIVA special issue; as well as Palmer and Reason 2009; Ziska et al 2013 for the observed based climatologies among others!

In this passage we also refer to the VSLS source gases in the tropopause region, as we are interested in the amount of bromine entering the stratosphere. The papers you mentioned are excellent studies of the source regions in the boundary layer, which is not the scope of our manuscript.

P. 4959, lines 25, 27 and 29: Add specific references here for the PG papers: Hossaini et al 2010, Liang et al 2010, Tegtmeier et al 2012. Here, no references are given at all!

References added as follows: “**The contribution of product gases (so called product gas injection) to stratospheric bromine thus needs to be determined either from modeling studies or from observations of source gases using specific assumptions on the transport into the stratosphere and chemical reactions and washout during this transport process (e.g. Aschmann et al., 2009; Hossaini et al., 2010; Liang et al., 2010, Tegtmeier et al 2012).**”

And please do not only refer to Montzka et al 2011 as a sum up for all available references. You need to give credit to the original publications as well as to new papers published after WMO (2011).

We checked the manuscript for more suitable and recent references then Montzka et al. 2011 and changed where appropriate.

P. 4960, L. 4: “these emission”. You were not talking about any emission before at all. In fact you are totally neglecting oceanic sources of VSLs and their emission! This should be added to the introduction as well. This is an important SHIVA outcome!

It is not in the scope of this paper to discuss the specific oceanic sources of VSLs.

“As mentioned above the sources of bromine VSLs to the atmosphere are largely of natural origin (e.g. Leedham et al., 2013). Consequently, the emissions of this species are expected to display significant geographical and temporal variability.”

P. 4960, L. 13-15: The “tropical” West Pacific is expected to be the most important source region...” The sentence is not correct, please rewrite. The referred publications analysed only the Tropical Tropopause Layer thus “the atmosphere above the tropical West Pacific”. In Aschmann et al 2009 they simulated VSLs transport from the upper troposphere into the stratosphere. None of these studies looked at the oceanic sources of the VSLs from the ocean surface to the stratosphere.

I do not refer to the oceanic sources or any specific substance in this context. The cited papers deal with general atmospheric dynamics and mass transport from the troposphere into the stratosphere. I refer to these transport processes of air masses. For better understanding, the text was changed to:

“The Western Pacific is expected to be the most important source region for air masses transported from the troposphere through the TTL into the stratosphere (e.g. Newell and Gould-Stewart, 1981; Gettelman et al., 2002; Fueglistaler et al., 2004; Aschmann et al., 2009; Fueglistaler et al., 2009).”

However, unique data exist for the tropical West Pacific and are already published by e.g. Butler et al 2006 JGR, Tegtmeier et al 2012 ACP, Krüger and Quack, 2013 ACP; HIPPO aircraft campaign (e.g. Tegtmeier et al 2013, ACP SHIVA special issue) and should be cited here as well.

At this point we only refer to the observation of elevated (> 5ppt of CHBr₃) mixing ratios. Neither Butler et al. 2006 nor in Tegtmeier et al. 2012 nor Brinckmann et al., 2012 reported on such exceptional high values. In the mentioned publications, maximum values of around 2.5 to 3 ppt CHBr₃ have been observed in the boundary layer.

Given that the SHIVA campaign included also a ship expedition, which was directly linked with the FALCON aircraft measurements, I totally miss the link between the aircraft and the ship measurements within this paper. You also need to cite other SHIVA relevant results for the VLSL sources in the coastal and open waters of the Malaysian waters/ South China Sea here (Leedham et al 2013 ACP SHIVA special issue; Fuhlbrügge et al in preparation), which are important for the interpretation of your own VLSL air measurements.

It is beyond the scope of the paper to investigate the different oceanic sources of VLSL and to directly link ship and aircraft measurements during SHIVA (see also general comment on this review).

Line 4960, Lines: 22-25: Not correct, please rewrite! Hossaini et al 2013 carried out chemical transport model simulations using different VLSL climatologies as input and compared these model simulations with NOAA/ESRL ground stations measurements. **“Hossaini et al. (2013) used their 3-D chemical transport model to evaluate a range of different emission inventories (Warwick et al., 2006; Liang et al., 2010; Saiz- Lopez et al., 2012; Ziska et al., 2013). For this purpose the model outputs for the different emission inventories were compared to independent atmospheric observations (long-term observations at National Oceanic and Atmospheric Administration (NOAA) ground-based stations, HIPPO and SHIVA aircraft campaigns), showing that significant differences in these scenarios exist for the Western Pacific region.”**

P. 4968-4969, L 18-4: How in detail was the BL height calculated or derived? This is totally unclear! How was it done for the radiosondes launched onboard of the ship in contrast to the aircraft data? There must be technical differences? Where were the radiosonde measurements carried out and where the aircraft data (new Fig. 1)? The statement citing Roedel reference does not fit for the convective active West Pacific, better delete! Since 2012 there is no IFM-GEOMAR anymore, now called GEOMAR.

There is no difference in the calculation of BL height between ship and aircraft measurements as both are using data of temperature and humidity. For clarification, we changed the text to:

To determine the thickness of the boundary layer during SHIVA, the profiles of potential temperature, relative humidity and wind speed as well as the bulk Richardson number are used to calculate an average value as described in detail in Fuhlbrügge et al. (2012).”

IFM-Geomar changed to GEOMAR.

Seibert et al. (2000) is used instead of Roedel (2011) now.

P. 4970, L. 4: What is with VLSL data between 450m and 2 km? You totally neglect them?

The data between 450 m and 2 km is not considered further in the discussion, because mixing between the boundary layer and the free troposphere is not subject of this paper. For further considerations of the mixing processes in this altitude range, detailed informations about the boundary layer have to be taken into account and this is beyond the scope of this paper.

P. 4971, L. 10-13: Exclusion of the high VLSL abundances for the flight 20111119a, but why? Could there not be high oceanic sources at the coast between Miri and Kuching? Please cross-link with oceanic measurements taken from the SHIVA campaign as well! Use the advantage of being a member of a multidisciplinary EU project to deliver more insights in this interesting field of science! In general, I believe, high priority should be given for a special issue journal to carefully cross link between different publications.

Having a look at unpublished data of the flask and μ -dirac data from the RV SONNE measurements of the 19.11.2011 reveal, that there are no elevated mixing ratios visible for CH₂Br₂. Unfortunately, CH₂BrCl has not been measured at the ship.

CH₂BrCl and CH₂Br₂ have also been measured by the WASP from UEA and the agreement with our measurements is very poor. This cast doubt on the integrity of these data and therefore we decided to reject them. See also answer to Referee #1 comment on the same topic.

Conclusions:

P 4985, L. 1-2 But why are the values higher than for Montzka et al 2011? Which areas did the Montzka et al/WMO 2010 data cover? What is different? Were any tropical West Pacific coastal and open data included? No, just recently the HIPPO aircraft and now ATTREX aircraft data are carried out for the tropical West Pacific. **As noted in the manuscript, the data is from the upper troposphere (10-13km), where the direct coastal influence is expected to be small. The only available typical mixing ratios from HIPPO in the tropical upper troposphere are presented in Wisher et al. (2013), covering only 7 data points and missing CHBrCl₂ and CH₂BrCl. Therefore we decided not to consider them. The data from the ATTREX campaign is not published yet.**

P. 4985, L. 20: rewrite: "This shows that the West Pacific may be a preferred...". Cut out "may" as we do have evidence for this (see my comments above) and ex-change "West-Pacific" with "Malaysian coastal and open waters" (see also my title comment above).

Please note that were are not referring to the VLSL emissions in the boundary layer. We are looking at the transport into stratosphere. This is - in contrast to the emissions in the boundary layer - not a local phenomenon. The tracer composition in the upper troposphere are determined by processes from local (convection) to mesoscale range (horizontal transport and mixing).

P. 4985, L. 23-26: Rewrite as there are a lot of new papers with VLSL measurements and modelling together showing clear evidence for the tropical West Pacific being the source region for oceanic VLSL into the stratosphere e.g. Warwick et al 2006; Tegtmeier et al 2012/2013, Hossaini et al. 2013.

We clearly state that more vertically resolved measurements are needed to confirm an upper tropospheric budget.

Table 6: Compare with other new publications. There are a lot of new VLSL data for the MBL available! Cite and use other available tropical West Pacific data from Pyle et al 2011, Brinckmann et al 2012, Ziska et al 2013, Hossaini et al 2013! Do not stick to the “older” WMO (2011) data.

Ziska is focused on emissions, only considering CHBr₃ and CH₂Br₂ and no typical atmospheric mixing ratios for the Western Pacific are given.

Atmospheric mixing ratios are compared in Hossaini et al. (2013), including our data from SHIVA. We included the measurements of Brinckmann et al. (2012) in the table, as those data covers all 5 VLSL and are on the same calibration scale as our measurements.

Table 8: Here you compare coastal and open Malaysian waters, an area with extensive convective activity, with VLSL measurements outside of the tropical West Pacific area! Clarify this in the corresponding text of the ms. This may explain the large differences between the two.

The intention of Table 8 is not to compare the Western Pacific with other regions, but to compare typical values of mixing ratios in the boundary layer and upper troposphere to see the decay between the two altitude intervals. For clarification we added the altitude interval used for the upper troposphere in the table heading.

Figures 2, 4, 5: The outliers for 20111119a may not be measurement problems, but may be due to elevated oceanic sources. Please check with oceanic measurements carried out within the SHIVA campaign and add this to the discussion of the paper.

As explained above, we cannot exclude instrumental problems, as simultaneous measurements from WASP do not support these high mixing ratios. Therefore we prefer not to consider these data in averages.

Minor and technical comments:

Whole ms: Three different usages of boundary layer abbreviations: PBL, MBL, BL, please synchronize.

Changed where appropriate to PBL (planetary boundary layer).

Whole ms: Montzka and Reiman et al 2011 should be exchanged with Montzka et al 2011.

Changed where appropriate.

P. 4958 Line 18: “all four halons” unclear which one you refer to here! Be specific.

Specified now.

“...including four halons (H-1301, H-1211, H-1202, H-2402), CH₃Br and the VLSL...”

P. 4959, Line 4 and P 4958 Line 13/14: Use the same abbreviation for VSLs.
Changed where appropriate to “very short-lived species”.

P. 4959, L. 16 Add “oceanic upwelling regions” as well.
“...in particular in coastal regions and oceanic upwelling regions (Quack and Wallace, 2004)”

P. 4959, L. 19: unclear, correlates with what?
See answer to Reviewer #1 comment.

P. 4960, L. 5: double use of “change”, rewrite.
“They are further prone to changes due to variations in climate or due to land and ocean use, e.g. seaweed farming.”

P. 4961, line 20: November-December is not “fall” anymore! Either write early winter or write out the month.
“As part of this project a field campaign in the Western Pacific was conducted in November and December 2011.”

P. 4964, l 2: Unclear, what is the sampling interval now? Every _7 min or every 4.3min?
The sampling interval is 4.3 min. Sentence clarified as follows: “...sample cycle of 4.3 minutes, including a chromatographic runtime of 2.9 minutes.”

P. 4965, L. 12: Unclear to me, what is the sampling interval for WASP? Every 20 to 180 s?
WASP did not sampled on a regular time interval, so there is no exact number for the sampling interval. The flushing time of the sample flasks was altitude dependent. It was varying between 20 and 180.

P. 4971, L. 21-22: Flight 20111119b was also probing convective outflow!
You are right, and also several other flights (e.g. 20111202a, 20111209b). We chose these two flights, because we probed the convective outflow over a very long flight distance.
“Especially two flights performed towards the end of the campaign (20111211a & b) probed over a long flight distance the outflow from several large convective cells at an altitude of approximately 11 km.”

P. 4973, L. 21-23: High atmospheric VSLs abundances due to high oceanic sources?
As explained above, we cannot exclude instrumental problems, as simultaneous measurements from WASP do not support these high mixing ratios. Therefore we prefer not to consider these data.

References:

Newell and Gould-Stewart: something is wrong with the reference details.
Changed in the references.

Oram et al 1995: write capital letters for HCFC.
Changed in the references.

Table 1; MBL stands for?

MBL stands for marine boundary layer. A legend for the abbreviations was added to the table caption.

Table 4 and 6: Shift the PBL data to the left and UT/ MBL to the right as described in the table caption.

For convenience, the table caption was changed to “Overview over the averaged mixing ratio of the VSLs in the upper and free troposphere and the planetary boundary layer.” in table 4.

I guess, you refer to table 5, not 6. In table 5, the caption was changes to “Overview over the mean mixing ratios of the long-lived halons and CH₃Br in the upper troposphere and the planetary boundary layer...”

Table 7: If possible compare also with HIPPO aircraft data! Clarify that Wisher et al 2013 is eq. CARIBIC data!

As explained above, the only available data from HIPPO in the tropics are presented in Wisher et al. (2013), covering only 7 data points and missing CHBrCl₂ and CH₂BrCl. Therefore we decided not to consider these data. „... data published in Wisher et al. (2013) from South East Asia (0-15°N), derived from CARIBIC measurements.”

Table 10: “of an individual substances” Sg or PI?

Changed to “of an individual source gas”.

Table 11: Which WMO data? Reference is missing here.

Reference (Montzka et al.) was given above in the table caption.

“...given in Montzka et al. (2011), denoted as WMO.”

Figure 2: Blue bars are not visible.

We think that the blue bars are well visible, however, for clarification we changed the text to “blue horizontal bars”.