We thank reviewer 2 for taking the time to make an extensive and thorough review. Based on the general comments of both reviewers, we felt it was necessary to restructure the paper in a major way. Below we give a summary of each of the reviewer's arguments and then we provide a general outline of these major changes to each general comment separately.

We have quoted excerpts of reviewer 2's comments below and they are shown in italics. Excerpts of the paper quoted by the reviewer are shown in double quotations and italics, and excerpts from the paper that we quote are in sans serif font and quotation marks. Modifications made to the article are shown in bold.

The reviewer has presented a series of arguments arguing that this work is not in anyway relevant to the stratospheric supply of bromine from VSLS. We present a summary of the reviewer's arguments and then respond to the general themes raised:

- 1) Air lofted above the level of zero radiative heating (LZRH), which is about 15 km in the tropics, will have a net tendency to ascend. Air lofted below this altitude will have a net tendency to descend.
- 2) The observations made by the Falcon and the relating modelling analysis occurred below the LZRH, and thus sampled air with a net tendency to descend.
- 3) Observations, and by extension, modelling analysis below the LZRH have no relevance to the stratospheric supply of bromine from VSLS. Thus, it is argued, this work has no relevance to the stratosphere.
- 4) We should consequently recast the paper purely as a discussion about the impact of VSLS and iodine compounds on tropospheric chemistry.

We first need to start by explaining why we think the reviewer made this criticism. We believe it is because we unfortunately failed to explain or justify a key idea. In short, that processes that chemically alter, emit, transport, or remove bromine in the troposphere can ultimately impact the eventual fluxes of bromine into the stratosphere. In full, CTMs and CCMs use a variety of simplifying assumptions regarding VSLS emissions, chemistry, washout, and transport. Here is a short list of the assumptions used in CTMs and CCMs that are relevant: the uniform distributions of VSLS in the boundary layer in place of emissions, the use of convective parameterisations, the use of simplified treatments of Br_v washout, and the use of reduced VSLS degradation schemes. These assumptions directly impact upon their representation of the bromine transport, speciation, and loss in the troposphere. Tropical deep convection is the primary means by which tropospheric gases are first lofted to the upper troposphere and above the level of radiative heating from where they may undergo eventual slow ascent to the stratosphere. Therefore, the assumptions regarding the tropospheric chemistry and transport of VSLS in global models can have indirect relevance to their estimates of strat-BryVSLS. We use a model that represents in detail the chemistry, transport, washout, and emissions of bromoform and its PGs to examine the tropospheric chemistry and the assumptions used in CTMs and CCMs. Such an approach therefore has an indirect relevance to the stratosphere. Finally, in point 4, the reviewer recommended that we remove all mention of the stratosphere and focus entirely on the troposphere. We have done this in so far as we have removed all discussion of direct transport and influence on the stratosphere and made it clear what our aims are.

Given that the current version of the manuscript lacks sufficient discussion of the reasoning above we have modified it in the abstract, introduction, aims, discussion, and conclusions to solve this problem. Note too that these improvements were also recommended by reviewer 1. This point relates directly to the main objectives of this work, so we have rewritten the objectives and include the new text below:

- To understand the chemistry and transport of CHBr3 and its PGs, and to estimate their chemical budget in the troposphere within an observed convective system.
- To discuss how these key processes relate to assumptions regarding the surface emissions, tropospheric chemistry, and transport of CHBr3 and its PGs within existing CTMs and CCMs.

Now, for clarity, we would also like to briefly address what this work did not seek to address and cannot. Due to the regional domain that we use, and the short timescale of the simulation (three days), we cannot simulate the transport and chemistry of VSLS and its PGs above the LZRH or make any estimates of strat-BryVSLS. Although not discussed in this work, VSLS and PGs that are transported up to levels below LZRH can be relevant to the stratosphere since subsequent convection can loft this material vertically.

The reviewer's second major comment focuses on various summarizing statements that we made. The reviewer argues that we have not fairly represented the true model to observation comparisons in these statements, and believes that they are inconsistent with one another. The reviewer mentions comparisons to the meteorology and also to the chemistry. Reviewer 1 also pointed out these apparent discrepancies, so we answer their comments here too. Rather than quote the entire argument from the reviewer here and respond in line we try to paraphrase it here in a series of numbered points. We then attempt to address each of these points in turn.

- 1) During Flight B, the modeled CHBr3 mixing ratios compare poorly to the observations from the aircraft. At various points in the paper we used this comparison as basis to argue, in summary, that the modeled CHBr3 in the convective system compared reasonably to observations. The reviewer highlighted these instances of summary as being problematic.
- 2) The comparisons between simulated and ship-based observed CHBr3 in the boundary layer did not support the mention of CHBr3 in the summarising statements either. Reviewer 2 indirectly referred to this point and it was directly raised by reviewer 1. Reviewer 2 also complained that that the mixing ratios from the second boat cruise (relevant to this point) did not match Table 4.
- 3) The reviewer argues that the comparison is even poorer for Flight A using the observations from both instruments on board the aircraft as a basis for arguing this.
- 4) The reviewer thought that our explanation of the emissions' performance was too long and was apparently unconvinced or did not understand some of our arguments.
- 5) Consequently, the reviewer did not agree with our statement that the simulated CHBr3 mixing ratios at Bukit Atur were consistent with the annual mean, but showed discrepancies during this simulation period during the Winter monsoon.

6) In a related point, the reviewer indicated belief that either the emissions were wrong or there was a problem with the meteorology.

Overall, we feel these criticisms are all symptoms of the same problem: we presented all of the available observations whether they had direct relevance to the convective cloud we that focused on or not. This had the consequence of making the paper very long, it forced us to make a lot of overly complicated explanations, it diluted the main points that we wished to address, and our summary statements were both not able to be concise and completely accurate at the same time. Our summary statements were accurate in relation to the observations supporting our conclusions, but it was not made clear that that they only related to these specific observations. In addition, some of our claims were not clearly enough described. This was particularly the case for the CHBr3 comparisons made during Flight B. In light of this we have removed many observations from the paper, and we have re-structured the paper to highlight how the observations support our conclusions. With regard to the key comparisons for CHBr₃ (see points 1 and 2 above) we have made some of the comparisons clearer. We describe these changes in more details below:

- We cut down figure 13 (now figure 12) and highlighted the observations made in the convective cloud and those outside of it. Consequently, we were more easily able to highlight the agreement between the model and the observations in the outflow plume.
- We have removed all of the observations made during Flight A since this flight did not observe convective activity and the observations were made in the boundary layer in a region that was not dynamically connected to the region near Flight B. This addresses the reviewer's third point.
- We have now structured the results and discussion according to the specific observations that we use to support different components of the case study, e.g., marine boundary layer observations, convective outflow observations, meteorological observations supporting general circulation, and meteorological observations relating to the studied convective cloud. This now makes the exposition of the results clearer.
- In particular, we highlight more clearly the marine boundary layer observations from the second local boat cruise and how they support our conclusions regarding the entrainment of air enriched with CHBr₃. These observations are now discussed in a specific section and the data are added to Table 4. We explain how they support our conclusions regarding the entrainment of CHBr₃ enriched air into the convective system.
- All of the CHBr3 observations aside from those made during Flight B are now shown only in either Table 4 or 5. The temporal variability of the other comparisons are not directly relevant to our conclusions.
- We have moved the discussion of the emissions to a new section, and we have reduced length of the explanation and have both simplified and clarified it. This addresses the reviewer's fourth, fifth and sixth points.



Here is a reproduction of the new figure comparing CHBr₃ during Flight B:

Figure 13. (a) This plot shows observed (black solid line) CHBr₃ and its accompanying 1σ error range (black dashed) in comparison with the simulated CHBr₃ (orange line) along the offset flight track. (b) The solid line shows the observed CHBr₃ variability both within the outflow plume (yellow shaded) and outside of it (grey shaded). The aircraft altitude is represented by the blue solid line in both figures. In addition to the precision of the observations, there is also an uncertainty due to the accuracy to which the standard is known that is equivalent to 4.5%.

We need to make several clarifications:

• The error bounds are equivalent to 1 sigma. The reviewer questioned this in their comments. We changed the paper to reflect this.

- This plot includes a more recent version of the GHOST-GCMS CHBr3 data. We regret that have changed the data subsequent to submitting it to ACPD, but this was due to revised post-processing of the GHOST-GCMS data. We are assured that this is the final version of the data. The new data has improved the model to observation comparison.
- In addition to the uncertainties shown in the plot there is an additional uncertainty due to the accuracy to which the standard is known. This uncertainty would act to scale all of the values either up down in unison. We have made this clear in the plot legends and text.

We have modified the paper to explain these key issues and to remove some of the confusion about what the data show.

The reviewer expressed similar concerns regarding our statements summarizing the meteorology. The reviewer highlighted the summarizing statements below from the paper indicating that the meteorological simulation during the case study was reasonable:

"Top of page 20631 states" "Having established that the simulation offers a sufficiently realistic representation of the meteorology during the case study ..."

The key question is whether the original wording "sufficiently realistic" was appropriate or not? We assert that it was, but acknowledge this was not altogether clear in the original version of the paper. The reviewer then highlighted various instances when the model did a poor job of simulating the meteorology. A unique response is required to each of these concerns, so we quote the text highlighted by the reviewer and deal with the specific concern regarding each point individually.

"whereas Figure 7 shows large differences in modeled and measured wind direction,"

The differences described by the reviewer are as a result of the model failing to represent the precise altitude at which the wind direction changes. We do not believe that these differences will affect our conclusions because they do not occur within the altitude range on which we focus, i.e., 0- 12 km.

"Figure 9 shows a troubling offset between observed and simulated T,"

We acknowledge that the performance of the model's meteorology is at fault during Flight A. The Region near Flight A was dynamically unconnected from the region covered by Flight B, so these problems do not affect our conclusions. Due to the same reason, we have removed the comparison with Flight A from the latest version of the paper.

"and Figure 2 shows the modeled origin of convection (Region 2B) originated over land whereas radar imagery shows the actual convection originated over water (Region 2A)."

We discussed the offset at length in the paper. We acknowledged this problem would only affect our conclusions if the CHBr3 levels in Region 2A were substantially different from those in Region 2B. We argued that they were similar in the paper, and, in addition, provided observational support for the simulated values at both locations. We therefore demonstrated that this problem would not affect our conclusions. We have tried to make this discussion clearer now in the paper.

We now deal with a series of specific comments made by the reviewer. We thank the reviewer for highlighting a series of omissions regarding details of the instruments that measured CHBr3.

"...there is no description of the GHOST instrument. In some places GHOST-MS is used; in other places GHOST GC-MS is used. Is it a MS or a GC-MS? Are other VSL bromocarbons measured? "

We have now made it clear that GHOST is a GC-MS. We have cited a paper describing the instrument. We have also now described that it did observe other VSLS.

"If so do the measurements of the other VSL bromocarbons support the sole focus on CHBr3?"

The focus solely on CHBr3 is a result of constraints placed by the modelling. It is simply not currently feasible to simulate other VSLS and their chemistry in a model with such high spatial resolution whilst considering other processes such as washout in detail.

"The WASP instrument is also not described. In Table 2 it is called WASP GC-MS, so I assume this is a gas chromatograph / mass spectrometer."

"Why is the altitude range of WASP limited ? (page 20633 states WASP can not make measurements for altitudes above 6 km)."

WASP is a GC-MS. Due to a fault, the WASP inlet system was not able to draw in sufficient air whilst at high altitude. Thus, the maximum altitude that they could measure was 6 km. We have actually removed the Flight A observations, and so no longer use the WASP data.

Next, the reviewer has indicated surprise that we represent microphysical processes in a model with a horizontal resolution of $2 \ge 2$ km. It is certainly true that we used a parameterisation in the model to represent the microphysics of cloud particles and hydrometeors. It would not be too strong a generalisation to say that there is microphysical parameterisation, in some form, in every prognostic weather prediction model. Thus, it is so in our case too. The objective of such a parameterisation is to take the prognostic thermodynamical and dynamical parameters in the model and to use them to determine the mixing ratios and size distributions of the various condensate particles. Given that the use of such schemes is essentially canon in the field of numerical weather prediction, we do not see that it is necessary to justify using a scheme like this in the model.

The reviewer questioned how we calculated photolysis rates within the model. We thank the reviewer for raising this issue. We have therefore added this text:

The photolysis rates are computed on-line in the model using the Fast-TUV radiative model (Tie et al., 2003). This is done in such a way as to consider the effects of clouds on photolysis rate in an interactive way. We used the absorption cross-sections from JPL (Sander et al., 2006) and IUPAC (Atkinson et al., 2007) for the different photochemically active compounds.

"1. Many key papers are not cited."

Add Saiz-Lopez, Salawitch, Ko and Poluet.

We have added citations to all of the papers recommended by reviewer 2 with one exception. We feel that reference to Iraci et al. 2005 seems inappropriate. It refers to experiments carried out in aerosols composed mostly of sulphuric acid at temperatures typical of the upper troposphere and lower stratosphere. Although our work in part concerns the upper troposphere, the reaction substrate for the HBr + HOBr reaction in our case consists of cloud particles and raindrops. This same point was noted in Marécal et al., 2012. Further, since we focus on a convective cloud with consequent heavy rainfall, sulphuric acid aerosol is scrubbed out of the ascending air.

The reviewer noted we had not properly discussed the Henry's Law constants that had been calculated in previous work. The paper cited in the methods section, Krysztofiak et al. discusses the Henry's Law constants. We have added a note that interested readers should pursue that article for more detail regarding those types of discussion.

The reviewer explained that we had mistakenly stated that bromoform had the shortest lifetime of any of the VSLS. We thank the reviewer for pointing out this error. We have now modified the manuscript accordingly to explain it has one of the shortest lifetimes.

"3. Line 13 to 15, page 20619, states "Thus, enhancements of up to 15 nmol mol-1 are therefore indicative of enhanced vertical transport.". Presumably authors mean enhancements of CO. If so, to me, such enhancements are indicative of entrainment of polluted air and not enhanced vertical transport!"

We thank the reviewer for pointing out this error. We have changed the language to indicate that the enhancements are indicative of entrainment of boundary layer air. Concentration gradients of this magnitude were consistent with the background vertical profiles observed by the aircraft during the campaign, so we feel that "pollution" is probably too strong a description in this case.

The reviewer expressed a lot of concern that we stated we used a retention coefficient of one in the model to describe how chemical species were lost in ice particles. The reviewer argued that this assumption needed to be justified, claimed that it was not justifiable in their opinion, and that it may account for the efficient washout of soluble species in the model. We thank the reviewer for highlighting their concern surrounding this assumption. In response we have changed the text in order to justify this assumption and to explain it further.

Finally, chemical species dissolved within cloud particles and raindrops that undergo freezing are considered to be lost. Thus, we assume a retention coefficient of 1 for all species indicating that all of the chemical species is transferred into the ice phase upon freezing. In a convective system, this loss is due to the transfer of soluble gases into the liquid phase in either cloud particles or rain drops that then subsequently undergo freezing to yield one of the types of frozen condensate considered in the model: snow, aggregates, graupel, and hail. In addition, we do not consider the uptake of soluble species onto ice particles once they have formed. We do this to reduce the computational burden of tracing the chemical and transport fate of the soluble bromine species amongst all of the different types of ice precipitate. This is justified for two reasons. First, our results are insensitive to this assumption. It does not strongly impact the total wash out of soluble chemical species transported vertically by convection from the lower troposphere such as soluble inorganic bromine species. This is because much of the wash out of these species is made by the rain droplets below the altitude where ice particles are formed. Second, it is a reasonable assumption

within a convective cloud. In our simulation, the majority of ice particles within the convective column are actively growing by riming. Ice particles that are simultaneously undergoing riming and are adsorbing soluble chemical species do not present a suitable substrate for chemical reactions that might eventually release reaction products into the gas phase. This is because chemical species adsorbed onto ice particles undergoing active depositional growth will become trapped and buried in the bulk ice. In addition, ice particles that have reached sufficient mass will sediment rapidly acting to quickly remove the soluble species. This leaves only ice particles that are both light enough not to sediment and those that are not undergoing active growth as a suitable substrate for reaction, which probably represents a relatively small proportion of the particles within the active part of the convective system. On the other hand, the outflow region consisting of ice cirrus presents a situation for ice particles that are both long lived and that are likely not undergoing active growth. Owing to their longevity they likely do not represent a large removal source of Br_{y} , which is consistent with Aschmann. Instead, they likely represent more a substrate for heterogeneous chemistry that could in turn lead to compositional changes in Br_{y} in the upper troposphere over longer periods of time.

The reviewer was concerned that we had described our model as being cloud-resolving even though it has a spatial resolution of 2 x 2 km in the finest scale grid. This is not a controversial usage of this term in this context as implied by the reviewer, and we think this comment has arisen due to a misunderstanding. "Cloud resolving model" (CRM) is a commonly accepted term describing models with horizontal resolutions of 4 km or lower that are capable of resolving precipitation in cloud systems at that scale. The paper by Khairoutdinov et al. (2005, AMS) gives an example of the use of CRM within a GCM and is titled "Simulations of the Atmospheric General Circulation Using a Cloud-Resolving Model as a Superparameterization of Physical Processes", which was the first hit on google scholar for this term is but one example of many papers using this term. The term simply means is that it resolves the dynamics explicitly at the cloud scale. In practice, this means that in such models the microphysics is explicit at the cloud scale and therefore the precipitation is also explicitly resolved and there is no need for a convective parameterization in order to produce precipitation.

The reviewer questioned how we calculated the mean and background values for CHBr3. We calculated the mean in each case by simply taking the mean average of all of the data for either the model or the observations. We calculated the background by taking either the lowest observed or simulated values from these different locations. We no longer use backgrounds and now quote ranges. We feel that the existing description of the mean is sufficient given the context. Unless the reviewer feels strongly about this we would like to keep it as it is.

The reviewer questioned whether we should in fact have used the measure of enhancement above background from both the observations and model. We actually performed some sensitivity tests not described in the manuscript where we scaled the background CHBr3 fields from TOMCAT. We found that the values of CHBr3 in the convective outflow plume itself were insensitive to these variations decreasing by at most 0.1 pptv. Unsurprisingly, the background was highly sensitive to these changes decreasing by at least 0.3 pptv. Thus, these tests seem to suggest that the current metrics appropriately highlight the problems with the background and the genuinely good model performance in the outflow plume. We have not explained this issue in the text and hope that this explanation provides adequate justification for these choices.

"7. Lines 17 to 26, page 20634. First, I doubt the discrepancy could be resolved by OH, but hard to assess without knowledge of impact of clouds on JCHBr3."

The discrepancy discussed line 17, page 20634 refers to a discrepancy in ozone since this is what is discussed in the sentence immediately prior. We were a bit confused by this statement because we do not claim that a discrepancy in ozone could be solved by OH. Perhaps the reviewer was referring to the discrepancies for CHBr₃? In fact, we are claiming that discrepancies in ozone may affect OH since the photolysis of ozone is the primary (as in initial) source of HO_x in the troposphere, and therefore OH, via reactions (1) and (2)

$(R1) O_3 + hv$	\rightarrow	$O^1D + O_2$
(R2) $O^1D + H_2O$	\rightarrow	20H
(R3) NO + HO_2	\rightarrow	$NO_2 + OH$

Further, over this region, there is significant depletion of NO_x in air masses arriving from the Pacific (Rex et al., 2013 ACPD this issue), which account for the majority of air reaching the troposphere over Borneo during the winter monsoon. In the absence of HO_x recycling via (3) the abundance of ozone and its processing via reactions (1) and (2) is therefore the likely limiting production term controlling OH abundances.

"Second, OH in the region of interest is likely dependent on the accuracy of modeled NOx, due to HO2+NO."

Rex et al. that is in review in ACPD in this special issue discusses the general characteristics of the troposphere over this region specifically with regard to OH sources. Rex et al. argue that most of the free tropospheric air advected over Borneo has undergone long-range transport from the central pacific region. They therefore argue that the air arriving over this area is likely depleted of NO_x and has only background levels of ozone throughout the entire depth of the troposphere. They argue specifically that ozone represents the main production source of OH in the absence of NO_x .

"Unless there is a comparison of measured and modeled NO, I place little weight on the evaluation of the modeled OH described in this section."

 $(\text{R4}) \text{HO}_2 + \text{HO}_2 + \text{M} \quad \rightarrow \qquad \text{H}_2\text{O}_2 + \text{O}_2$

Knowing more about NO_x would certainly tell us more about the recycling that occurs between HO and HO_2 and the extent to which losses of HO_x via reaction (4) to produce H_2O_2 are prevented. However, given that we actually make a comparison with observations of OH and OH climatologies from this region we feel this bears more weight than an examination of HO_2/OH recycling, especially given its apparent likely insignificance in this region.

"8. Appendix gives a list of reactions. Sorry, but need rate constants (or origin of rate constants) for this to be suitably documented. Most importantly for the three Aqueous Phase reactions, need to know reaction probabilities!"

We thank the reviewer for highlighting that the paper lacks sufficient detail regarding the chemical mechanism. We have now changed the text to make it clear that the details of the chemical reaction rates can be found in the appendix. The details of where to find the reaction rates are now described as follows in the appendix:

We present the list of the chemical reactions in the RELASH mechanism. The RELASH mechanism is derived from the ReLACS scheme developed by Crassier et al., 2000, and we added halogen chemistry consistent with that developed by Hossaini et al., 2010 and Krysztofiak et al., 2012 for SHIVA. All of the reaction rates associated with the ReLACS scheme are described in Stockwell et al., 1997 in table 2. The subsequent modifications to the NMHC chemistry reactions rates to move from RACM to ReLACS are described by Crassier et al., 2000 and listed in table 3. All of the halogen and VSLS gas phase chemical reaction rates unique to RELASH are described by Krysztofiak et al., 2012 in table 4 of that article. Finally, the reaction rates for the aqueous phase halogen chemistry listed as unique to RELASH are described in detail within Marécal et al., 2012. Where appropriate specific details for the location of the detailed description are given.