

We thank both reviewers for their constructive comments. Our responses are given below in red text. Our modifications to the manuscript text are also highlighted in red for ease of identification.

Response to anonymous referee #1

Suggested changes/comments:

Page 1922, line 25: “On balance it seems likely that the short-lived bromocarbons are responsible for the apparent deficit.” It should be more clearly stated whether there is consensus among the referenced authors that the short-lived bromocarbons are responsible for the deficit or if this is the summation of the authors here.

We have modified this sentence to:

‘and overall it seems likely that the short-lived bromocarbons are responsible for the apparent deficit’.

Page 1924, lines 4-6: Delete the sentences “In Sect. 2.1 we briefly describe the instruments used for this work and discuss how we monitored instrument performance throughout the deployment. In Sect. 2.2 we discuss the calibration methodology used for each instrument.” This summary of the methodology section is not necessary, the titles make this clear enough.

We have removed the two relevant sentences.

Page 1924, line 12: “Briefly, each sample is pre-concentrated using a Carboxen trap.” Specifically which Carboxen adsorbent is used (mesh size etc)?

We have modified this sentence to:

‘Briefly, each sample is pre-concentrated using a dual-bed adsorbent trap (1 mg Carboxen-1016 60/80 mesh followed by 1 mg Carboxen-569 20/45 mesh)’.

Page 1925, lines 3-9: It should be clarified how many sites the instruments were operated at Bukit Atur, Danum and Tawau are all mentioned here yet only Bukit Atur and Tawau are described in more detail in the field deployment section on pages 1927-1929

On line 5 ‘Danum’ should say ‘Tawau’ (now changed in text).

Page 1926, line 6: “are always completely retained on the lighter Carboxen at the front of the adsorbent bed” Is this a multi-adsorbent trap? If so this should be stated in the brief instrument description given earlier along with the type of Carboxens used (see earlier comment).

Yes, it is a dual-bed adsorbent trap, we have now clarified this on page 1924, line 12 and mention here that ‘the weaker absorbing Carboxen (Carboxen-1016)’.

Page 1932, line 21 onwards: Discussion of the discrepancy between measurements of perchloroethene at the Bukit Atur and Tawau sites during the first few months of instrument deployment is explained by a drift on the calibration gas used at the Bukit Atur site. This does indeed appear to be the most likely explanation for the differences observed, however, little text is devoted to these differences within the text when the time-series and seasonal averages are explained. The text should make it clear that the perchloroethene reported at Bukit Atur between December and March is likely an upper estimate of the actual mixing ratios at that time. This should be reiterated when

describing the seasonality in figure 8 otherwise the text and plots could be considered misleading to the reader.

A fair comment: we have added additional text on page 1933 to re-iterate that drift in the calibration gas at Bukit Atur is the most likely cause of the discrepancy in perchloroethene, compared to Tawau where we had a higher quality calibration cylinder.

Pages 1937 line 24 to page 1938 to line 7: During discussion of correlation between  $\text{CH}_2\text{Br}_2^*$  and  $\text{CHBr}_3$  varying between months/different periods the authors should mention the possibility of interference from a changing  $\text{CHBrCl}_2$  component of the  $\text{CH}_2\text{Br}_2^*$  signal. If this is not thought to give any significant interference then it should be stated why in this section.

While we cannot rule out a significant interference in the  $\text{CH}_2\text{Br}_2^*$  signal from a changing  $\text{CHBrCl}_2$  component in these data, we believe that the contribution is minor and now refer in the text to other work where this has been the case. We feel justified in saying this because recent preliminary data from Tawau, where we are able to achieve a partial separation of  $\text{CHBrCl}_2$  and  $\text{CH}_2\text{Br}_2$ , always show the  $\text{CHBrCl}_2$  peak to be the minor component (10-30% of the peak height of  $\text{CH}_2\text{Br}_2$ ). This is true at other southeast Asian locations as well. We also now report in the text that the ECD is, per molecule, less sensitive to  $\text{CHBrCl}_2$  than to  $\text{CH}_2\text{Br}_2$  which has the effect of reducing the overestimation of  $\text{CH}_2\text{Br}_2$  by the use of  $\text{CH}_2\text{Br}_2^*$ . See reply to referee #2 for change in text.

Page 1938, section “5.4 Methyl iodide”: In previous sections where other compounds measured have been discussed, an approximate atmospheric lifetime of each compound was given, this would be useful here, making the sections more consistent and help the reader to better interpret the results.

A fair comment: we have added an atmospheric lifetime for  $\text{CH}_3\text{I}$  to the first paragraph of section 5.4, taken from the WMO Scientific Assessment of Ozone Depletion:2010 (also shown in Table 1 of our paper).

Page 1940, line 20 onwards: The authors should exercise care here and consider rewriting some of the text. The fact that the larger variability observed at the coastal site is not captured by the model is ultimately a short-fall of the model and its insufficient resolution. The better comparison of measurement and model at the inland site is indeed encouraging, but the differences at the coastal site are equally important and shouldn't be disregarded. A sentence or two to clarify this would be worthwhile here.

Please see response to referee 2.

Page 1941, line 6: The authors introduce the acronym VSLS without definition. While this term is common in the field it should be defined before first use in the text.

We have modified the sentence starting on Page 1941, line 4 to:  
'There have been somewhat conflicting reports of the fraction of Br in  $\text{CHBr}_3$  and  $\text{CH}_2\text{Br}_2$  compared to the minor very short-lived substances (VSLS).'

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Response to anonymous referee #2

Suggested changes/comments:

Page 1924 line 12 Include which Carboxene type trap is being used with the instrument

Referee #1 also had the same comment, we have modified the text to:

‘Briefly, each sample is pre-concentrated using a dual-bed adsorbent trap (1 mg Carboxen-1016 60/80 mesh followed by 1 mg Carboxen-569 20/45 mesh)’.

Page 1925 paragraph beginning line 17 In this paragraph the authors discuss the coelution of  $\text{CH}_2\text{Br}_2$  and  $\text{CHBrCl}_2$ , and the discussion to report  $\text{CH}_2\text{Br}_2$  as  $\text{CH}_2\text{Br}_2^*$  without correction. The reasoning for this is solid, however, inclusion of a short paragraph on the likely hood of similar sources of  $\text{CHBrCl}_2$  and  $\text{CH}_2\text{Br}_2$  as well as the atmospheric lifetime would be good as this is included for the primary species of interest in the result section and would aid understanding the the  $\text{CH}_2\text{Br}_2$  section.

Fair comment. We have significantly modified the 3<sup>rd</sup> paragraph in section 2.1 to more fully cover the issue of  $\text{CHBrCl}_2$  co-eluting with  $\text{CH}_2\text{Br}_2$  and the extent of the likely overestimation of  $\text{CH}_2\text{Br}_2$  based on the use of the  $\text{CH}_2\text{Br}_2^*$  measurement. We have also extended the 2<sup>nd</sup> paragraph in section 5.3 to include a discussion on the likely similar sources and relative emission ratios of  $\text{CHBrCl}_2$  and  $\text{CH}_2\text{Br}_2$ . Finally, in section 5.5 we include new text (2<sup>nd</sup> paragraph) which discusses the likely impact on the total bromine estimate from  $[\text{CHBr}_3 \times 3] + [\text{CH}_2\text{Br}_2^* \times 2]$  which might be expected from a variable amount of  $\text{CHBrCl}_2$  present in the  $\text{CH}_2\text{Br}_2^*$  peak.

Page 1926 line 9 Would the authors please clarify what is meant by background air.

Agreed. We have modified this as follows:

‘Samples are calibrated by running frequent chromatograms from cylinders of clean natural air, previously filled at Niwot Ridge (Colorado, USA), at an elevation of 3.5 km above sea level. The cylinder air is subsequently enriched with known concentrations of the target compounds.’

Page 1936 line 17 "The observed time series..." This should be reworded such that the model is compared to the observations rather than how well the observations match the model.

We take the point, but think we already say this.

Page 1937 line 12 The discussion of the differences observed between Tawau and Bukit Atur mention the inclusion of up to 30%  $\text{CHBrCl}_2$  but not whether the authors would expect this ratio to vary with transport and consequently whether the differences between the two sites are due to dilution or differential processing of the bromocarbon within the airmass.

$\text{CH}_2\text{Br}_2$  and  $\text{CHBrCl}_2$  do have different atmospheric lifetimes (123 and 78 days respectively) which would imply that differential processing can play a role in any difference in  $\text{CH}_2\text{Br}_2^*$  measured between the two sites. However, in this case, we do not expect this to contribute to any measurable differences in  $\text{CH}_2\text{Br}_2^*$  as in terms of atmospheric transport, the sites are only a day or so apart (Matt to check). What may be more important here is the extent to which boundary layer air at Bukit Atur has

been diluted with free tropospheric air with a lower ratio of  $\text{CHBr}_2\text{Cl}$  to  $\text{CH}_2\text{Br}_2$ , given that the site is 426 m above sea level.

We will add a couple of sentences to the text to this effect.

Page 1940 line 20 Again, this needs rewording to indicate that the model needs to capture the observed data rather than the other way round. The fact that the resolution of the model prohibits it from capturing the high coastal values could be discussed in this light.

Actually, this sentence discusses the agreement just in the observations of bromine from  $[\text{CHBr}_3 \times 3] + [\text{CH}_2\text{Br}_2^* \times 2]$  at Bukit Atur and Tawau as presented in figure 8. We don't present a plot of modelled bromine versus observations here. We have modified the sentence on page 1940 line 22 to address the model resolution point:

'In fact, as the model we used here was unable to capture the large variability in the observations, presumably due to insufficient information on the spatial and temporal inhomogeneity of the emissions seen at the coast and to model resolution, we suggest that, for evaluating models, an inland site is preferable to a coastal site.'

Page 1941 line 4 "There have been..." This is the first use of VLSL and needs to be defined as although it is a relatively common term within the community there are variations.

Referee #2 also had the same comment, we have modified the sentence starting on Page 1941, line 4 to:

'There have been somewhat conflicting reports of the fraction of Br in  $\text{CHBr}_3$  and  $\text{CH}_2\text{Br}_2$  compared to the minor very short-lived substances (VLSL).'