

Interactive comment on “The contribution of natural and anthropogenic very short-lived species to stratospheric bromine” *by* R. Hossaini et al.

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Response to Referee 3:

We thank Referee 3 for his/her comments. The comments are repeated below (in *italics*) and our responses are given in **bold** text.

General comments: The paper uses a 3-D CTM to estimate the contribution of 9 VSLs to total stratospheric bromine. It partly builds on a previous study (Hossaini et al., 2010) by the main authors and compares modelled profiles of brominated VSLs with recent aircraft observations in the tropics. This paper is well structured

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and the data and results are presented clearly in the figures and tables. The results certainly add to the ongoing discussion about the role VSLs play for total stratospheric bromine. The work is suitable for publication in ACP and I recommend publication after addressing the following minor comments.

Specific comments: Page 23860, line 3 and line 25, and title: WMO ozone assessments 2007 and 2011 use 'substances' instead of 'species' for VSLs.

Ok yes, we will change 'species' to 'substances' throughout the text in order to be consistent with the WMO reports.

Page 23862, line 6/7: Where and when have the Laube et al. Measurements been taken? I guess that is important to repeat here.

Yes we will include this in the revised manuscript. These balloon-borne measurements were made in 2005 in the tropics (Teresina, Brazil, 5°04'S, 42°52'W).

Page 23862, line 13 to 15: Can you provide some more details on the HIPPO-1 campaign and the NOAA measurement programs. Probably most readers are not familiar with them.

Yes, in the revised manuscript we will include a paragraph describing the HIPPO-1 campaign and also the NOAA program.

Page 23863, line 21/22: Why don't you provide the chemical formulas of the halons like for all other species, instead of writing the abbreviation (H1211 etc.) in brackets?

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Ok, we will do this in the revised manuscript.

Page 23865, line 13 to 15: Can you provide longitude and latitude for these stations. That would help the reader.

Yes we will include the location of these stations in the revised manuscript.

Page 23869, line 23 and around this paragraph: Do you have an error estimate for your range of 4.9-5.2 ppt? Did you perform sensitivity runs to investigate the main sources of error in your model? Or could you just list the main uncertainties and in which way they would probably influence your result. Instead of just saying on page 23871, line 16 to 18, that more measurements of VLS are needed, you then could maybe also suggest, which meteorological parameters or other observations (or even laboratory measurements) would be most useful to improve your model predictions.

Assessing the absolute error on these model runs is challenging. Diagnosing convection in an offline model is one source of uncertainty and this is the reason we perform simulations with the online diagnosed convection and also with the offline archived convection. Another common uncertainty in these type of simulations arises from the model OH field. OH observations are sparse however comparison of OH profiles from the 1999 PEM TROPICS-B campaign with modelled profiles (not shown), show reasonable agreement. The model OH field in this case was found to be within the min-max variability of the available observations from the surface to 200 mb. Further error on our conclusions may arise due to error on reported rate constants for the source gas + OH loss reactions. In the revised manuscript we will include discussion on these and other potential sources of error.

Yes, in the revised manuscript we will include clear text on what other

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measurements/products would be desirable in the future to improve these simulations.

Page 23869, line 25: What type of measurement is the Dorf et al. (2008) study based on, and where and when were the data taken. That might explain why you compare your results with this study and not any others?

The Dorf et al. (2008) study used balloon-borne DOAS (Differential Optical Absorption Spectroscopy) measurements of BrO in the stratosphere. Combined with modelling work the authors were able to infer a VSLs Br_y contribution (from the BrO/Br_y ratio) of 5.2 (±2.5) ppt in the stratosphere. The study launched balloons over tropical Teresina (Brazil, 5°04'S, 42° 52'W). We will include a clear discussion of the Dorf et al. paper in the revised manuscript. Also, we will compare our results with other recent observation-based estimates of Br_y^{VSLs}.

Page 23876, Table 2: The lifetimes of CH₂Br₂, CH₂BrCl and EDB that you provide in table 2 are longer than 6 months. VSLs are by definition substances with a lifetime shorter than 6 months, WMO (2007) and (2011). Do we need a new classification, or are we at a point where we can say that these substances should be grouped together with CH₃Br and the halons? Can you comment on that.

Yes, our results show the local photochemical lifetime of certain VSLs can be longer than 6 months, particularly in the cold upper troposphere where the OH sink becomes slow (for species whose dominant loss channel is via OH). The species weighted mean lifetime in table 2 show CH₂Br₂, CH₂BrCl and EDB have lifetimes > than 6 months and thus according to these results are not 'very short-lived species' by the WMO definition. We will point this out in our revised paper. However, any discussion on changing the definition or groupings of

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halocarbons needs to be done in a forum such as the WMO assessment process.

Technical comments:

- 1. Page 23859: Add place after NOAA in the 4th affiliation.*
- 2. Page 23861, line 26: Should be Hoyle et al., 2011 (instead of 2010), like in the reference list.*
- 3. Page 23863, line 6 and 13: ERA-interim: use capital letters.*
- 4. Page 23865, line 28, page 23866, line 3 and reference list: It should be Montzka and Reimann et al. (2011) and not 2010.*
- 5. Page 23867, line 8: Remove one 'the'.*
- 6. Page 23867, line 10: Remove the 'with'.*
- 7. Page 23868, line 8: Remove one 'to'.*
- 8. Page 23868, line 11: You need to define SGI, since it has not been mentioned before.*
- 9. Page 23869, line 12: '... than the our....' - remove 'the' .*
- 10. Page 23872, line 24 to 26: Remove the Gettelman et al. reference, since it is not used in the text.*
- 11. Page 23877, Fig. 1: Enlarge Figure 1 for better readability.*

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12. Page 23879, Fig. 3: Enlarge Figure 3 if possible, or at least the axis labels for better readability.

13. Page 23882, Fig. 6: Enlarge tick marks, since they are overlaid by the lines in the bottom of the plot.

14. Fig. 2, Fig. 5, Fig. 6: 'mb' as a unit is not wrong, but I think it is more common to use hPa, or mbar.

Ok, we will address each of these technical corrections in the revised manuscript.

References:

Dorf, M., Butz, A., Camy-Peyret, C., Chipperfield, M. P., Kritten, L., and Pfeilsticker, K.: Bromine in the tropical troposphere and stratosphere as derived from balloon-borne BrO observations, *Atmos. Chem. Phys.*, 8, 7265–7271, doi:10.5194/acp-8-7265-2008, 2008.

Hossaini, R., Chipperfield, M. P., Monge-Sanz, B. M., Richards, N. A. D., Atlas, E., and Blake, D. R. (2010): Bromoform and dibromomethane in the tropics: a 3-D model study of chemistry and transport, *Atmos. Chem. Phys.*, 10, 719–735, doi:10.5194/acp-10-719-2010.

Laube, J. C., Engel, A., Bönisch, H., Möbius, T., Worton, D. R., Sturges, W. T., Grunow, K., and Schmidt, U.: Contribution of very short-lived organic substances to stratospheric chlorine and bromine in the tropics – a case study, *Atmos. Chem. Phys.*, 8, 7325–7334, doi:10.5194/acp-8-7325-2008, 2008.

WMO (2007), Scientific assessment of ozone depletion: 2006, World Meteorological Organization Global Ozone Research and Monitoring Project, Report 50.

WMO (2011), Scientific assessment of ozone depletion: 2010, World Meteorological Organization Global Ozone Research and Monitoring Project, Report 52.

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