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Atmospheric Chemistry and Physics Discussions

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 1:
We thank Referee 1 for his/her comments. The comments are repeated below (in *italics*) and our responses are given in **bold** text.

This concise, well-structured, and well-written paper presents a global three-dimensional chemical transport modelling study on the impact and contribution of 9 very short-lived species (VSLS) on the bromine budget of the stratosphere. In addition to major VSLS like $CHBr_3$ and CH_2Br_2, poorly studied anthropogenic VSLS

like C_2H_5Br, CH_2BrCH_2Br, n-C_3H_7Br, and i-C_3H_7Br have been also considered. Modelled tropical profiles of long-lived sources gases and a range of VSLS have been compared to the 2009 NSF HIPPO-1 aircraft campaign and ongoing NOAA measurement programs data and a reasonably good agreement is obtained. The modelled contribution of the VSLS to the stratospheric Br_y is found to be in the 4.9-5.2 range, in agreement with balloon DOAS observations. This study is a valuable contribution to the atmospheric bromine issue which is a timely subject matter. Therefore I recommend the paper for publication in ACP after addressing the following comments:

Specific comments:

Page 23861, line 28: Are nPB and iPB widely used and for which purpose? Please mention it.

nPB is currently used as a replacement compound for CFC-113 and CH_3CCl_3. It is used for industrial metal degreasing and electronic part cleaning (WMO, 2010). Similarly, iPB is a solvent and is also used industrially. We will include extra text with these points in the revised manuscript.

Page 23862, lines 13-14: In addition to references, a short description of the HIPPO-1 campaign and NOAA VSLS measurement programs is needed. That could help the readers who are not familiar with these data sets.

Ok, we will include a short description of the overall HIPPO-1 campaign.

Page 23864, line 15: The evaluation no 15 of the JPL compilation is used for rate constants and absorption cross sections but a new evaluation (no 17) has been recently released. Are there major changes between evaluations 15 and 17 which can have an impact on your modelling study?

No. The rate constants for the very short-lived source gases +OH sink reactions are unchanged between the two evaluations. The recommended absorption cross section data used for photolysis calculations are also unchanged.

Page 23867, lines 18-20: The choice of convection scheme is important for this kind of study. Do you expect a significantly different contribution of the VSLS to stratospheric Br_y if another parameterisation of cumulus convection was used in SLIMCAT?

We would expect some variation depending on the choice of cumulus convection. Parameterisations providing more frequent/intense convective upwelling would increase the modelled SGI. However, as removal of soluble Br_y is coupled to convective precipitation this could lead to a reduced delivery of Br_y from VSLS reaching the stratosphere via the PGI route. We will discuss this in the revised manuscript.

Page 23868, lines 5-6: C_2H_5Br and EDB are minor VSLS but due to their long lifetime in the TTL, they are potentially important bromine carriers to the stratosphere. Do we expect an increase of these anthropogenic VSLS in the future?

This is currently difficult to quantify as present day global emissions are poorly constrained. EDB is presently used for a number of different purposes (e.g., industrial fumigation) and C_2H_5Br is used as an ethylating agent, refrigerant, and a flame retardant (e.g., Low et al., 2003). Atmospheric observations of these species will help determine any potential growth.

Page 23869, lines 23-25: The modelled total Br_y/VSLS supply is of 4.9-5.2 ppt, which is consistent with the Dorf et al (2008)'s estimate inferred from balloon DOAS

observations of BrO. It should be noted that this supply of 4.9-5.2 ppt is somewhat larger than the values derived from balloon and aircraft measurements of the source gases in the TTL (0.7-3.4 ppt; WMO, 2010). Please comment this point.

Yes. The inferred range of 0.7-3.4 ppt is based on the source gases only – i.e., the SGI component of the total bromine delivered by VSLS. Inclusion of the product gas contribution (PGI) leads to the inferred range of 1-8 ppt for the total bromine supply (WMO, 2010). Our modelled total contribution (4.9-5.2 ppt) includes PGI and is within this range.

Page 23870, lines 13-14: Results are sensitive to the model OH field. Has this field been assessed through comparison to observations?

For this study we use monthly averaged 24-hour mean values of [OH]. Tropospheric variability in OH is large and 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. However, we also acknowledge that the model [OH] is uncertain. We will include discussion on this potential uncertainty in the revised manuscript.

Technical comments:

Page 23863, lines 20-22: The reference to Table 1 is not clear. You could replace the sentence by "In this study we include 9 bromine-containing VSLS SG tracers (see Table 1) and the following long-lived species: methyl bromide . . .".

Ok, we will make this clearer.

Figs 1 and 2: It is sometimes difficult to distinguish the Sdiagnosed (black line) and Sarchived (dark blue line) curves. The use of a lighter color for Sarchived would help.

Ok, we will use a lighter colour.

Fig 2: Please also indicate the TTL with dashed lines as in other plots.

Ok we will do 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.

Low, J. C., Wang, N. Y., Williams, J., and Cicerone, R. J.: Measurements of ambient atmospheric C_2H_5Cl and other ethyl and methyl halides at coasta l California sites and over the Pacific Ocean, J. Geophys. Res., 108 (D19), 4608, doi:10.1029/2003JD003620, 2003.

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

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 23859, 2011.