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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Bridging the gap between bromocarbon oceanic emissions and upper air concentrations" *by* S. Tegtmeier et al.

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In their manuscript, Tegtmeier et al. present emission fluxes of the two very short-lived brominated source gases (VSLS) CHBr3 and CH2Br2 from the Transbrom-Sonne shipborne campaign in the West Pacific together with calculations from a state-of-the-art particle dispersion model (FLEXPART). Modelled VSLS mixing ratios based on the observed emission fluxes are compared with previous observations in the tropopause region.

The manuscript addresses an important topic and has the potential to make a significant contribution to reducing current uncertainties in this field. However, I have a number of issues that in my point of view need to be addressed before this manuscript can be published.

1) As Ross Salawitch in his review has already addressed, uptake, sedimentation and heterogenous chemistry on ice in the tropopause region is a critical process controlling the input of VSLS product gases into the stratosphere. In Aschmann et al. (2011) we have implemented a representation of these processes in a global chemical transport model and found that sedimentation of inorganic bromine from the tropopause region is negligible, partly because of heterogenous chemistry that releases adsorbed bromine back into the gas phase. Consequently, Aschmann et al. (2011) found that almost all of the bromine from VSLS can enter the stratosphere. This appears to be in contrast to the findings of the current manuscript of Tegtmeier et al. It would be helpful to confirm or revise the results of Aschmann et al., but I'm not sure if the current study can provide more insight into the role of heterogenous chemistry ice uptake and sedimentation. One solution could be to acknowledge the uncertainty in this area and to put less emphasis on the calculated product gas injection and stress more the other aspects of this work. Aschmann et al. (2011) concluded: "If removal of Bry by uptake and sedimentation is indeed not very efficient then the flux of VSLS bromine into the stratosphere will be largely controlled by the distribution of VSLS in the troposphere and the pathways by which VSLS enters deep convective uplift"; this is precisely the area were the current manuscript of Tegtmeier et al. makes an important contribution.

2) At many places I found it hard to compare the results presented in the current manuscript with results from previous work. (a) It was not at all clear to me how to compare the calculated VSLS profiles given in units of nano mole (Fig. 2) with atmospheric observations. The description in Sect. 2.5 of how this was converted into mixing ratio profiles was rather confusing to me. While Fig. 2 shows the calculated profiles for the whole range from the ground up to 20km in moles, the mixing ratio profiles (Fig. 5) are shown only for the tropopause region. Is there a specific reason why? (b) Fluxes of VSLS (in nmol) are shown in Fig. 3, but unfortunately no corresponding atmospheric mixing ratios. Are these the same as shown in Brinckmann et al. (2012)? (Brinckmann et al. should be cited!) How well do the calculated VSLS amounts in the atmospheric boundary layer compare with observed concentrations? If I understand it

ACPD
12, C1087–C1090, 2012

correctly, emission fluxes are given in 10 nmol/hour/500m2 ? But is this also true for fluxes into the stratosphere (called entrainment here)?

3) Including estimates of the Ozone Depletion Potentials (ODP) adds to the study. However, currently this stands somewhat disconnected from the rest of the paper. I had difficulties to understand how the ODPs were actually calculated here. Were they calculated by using a different set of trajectories in the troposphere as the FLEXPART calculations used in the rest of the manuscript? If yes, how sensitive would the results be to the transport calculation? I would assume that the ODP for VSLS depends critically on the representation of (tropospheric) transport?

4) It should be made clearer what observations of VSLS in the tropopause region were used for comparisons and where potential limitations of this approach are (comparing data for different places at different times, etc.). The statement in the abstract that this study provides "the first link between observed oceanic emissions and in situ TTL measurements" raises false expectations and should better be modified.

I believe that most of these issues can be addressed by modifications to the text (and Figures) without the need to perform additional model calculations. With this I would hope that this study can make an important statement on the flux of VSLS into the upper troposphere, TTL and stratosphere based on observed ocean-to-atmosphere fluxes and state-of-the-art modelling in the important West Pacific region.

Additional references

Aschmann, J., Sinnhuber, B.-M., Chipperfield, M. P., Hossaini, R., Impact of deep convection and dehydration on bromine loading in the upper troposphere and lower stratosphere, Atmos. Chem. Phys., 11, 2671–2687, 2011.

Brinckmann, S. Engel, A., Bönisch, H., Quack, B., Atlas, E., Short-lived brominated hydrocarbons – observations in the source regions and the tropical tropopause layer, Atmos. Chem. Phys., 12, 1213–1228, 2012.

ACPD

12, C1087–C1090, 2012

Typographic corrections:

References: Carpenter and Liss: "Bromoform" should be lower case; Forster et al (2007): titel should be lower case; McLinden et al.: remove "." after "Sinnhuber"

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