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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Convective transport of very-short-lived bromocarbons to the stratosphere" *by* Q. Liang et al.

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The manuscript by Liang et al. investigates the delivery of bromine from very short-lived substances (VSLS) into the stratosphere. The main conclusion is that product gas injection (PGI) could be much larger than deduced from previous studies, so that the overall contribution of VSLS to stratospheric bromine would be much larger. This is an important finding that has implications for our general understanding of stratospheric bromine loading. The manuscript is well written and should be published in ACP after consideration of the following comments.

However, I have two general comments that I feel need to be addressed before publication of this study:

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Interactive Discussion

Discussion Paper

1. A main conclusion of this study is the finding, that product gas injection (PGI) could be much larger than deduced from previous studies. As is stated in the manuscript, an explicit treatment of bromine chemistry and the processes leading to its washout are critical to nailing down the estimate of stratospheric Bry from VSLS. However, on p. 655, l. 22 it is stated that bromine chemistry below the tropopause is not explicitely calculated here, but prescribed, with 80% of Bry being in the form of HBr and 20% as HOBr. What about the TTL? Is chemistry calculated in the TTL or only above the tropical cold point tropopause? If chemistry is indeed only calculated above the cold point, I'm having difficulties in understanding the differences in PGI between the present study and that of Liang et al. (2010).

2. The only information on PGI from atmospheric measurements comes from BrO observations. Recently, Kreycy et al. (2013) suggested, using balloon borne DOAS BrO observations, that the ratio of J(BrONO2)/k(BrO+NO2) should be much larger (factor $\sim$1.7) than calculated from current recommendations, which would have a significant effect on the calculated BrO/Bry ratio. I assume this was not considered here? A comment how this would affect the comparison with BrO here would be useful. On a different, but related point: The present study considers only the two most abundant VSLS CH2Br2 and CHBr3. Observations indicate that the minor VSLS such as CH2BrCl, CHBr2Cl and CHBrCl2 together contribute another $\sim$1ppt, which would need to be taken into account when comparing with BrO observations.

Specific comments:

p.653, l.6: should we really call this a "good agreement" given the large range of values (1-8ppt)?

p.653, l.24, somewhat misleading, as Brinckmann et al. is not a modelling study

p.655, l.9: Is the RAS convection parameterization different from the convective parametrization in the underlying GEOS-5 model? Would be good to provide a bit more detail.

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p.655, l.22: As noted above, this simple treatment is certainly not ideal when you want to investigate the fate of product gases. At least a caveat should be included and a more detailed specification how the tropopause is defined in the tropics. If chemistry is interactively calculated throughout the TTL this would be less critical than if chemistry is calculated only above the cold point.

p.656, l.4: Aschmann et al. (2011) showed that heterogenous reactions in the TTL have a large impact on bromine partitioning

p.658, l.5: Again, is bromine chemistry modelled below the tropical tropopause (cold point) or specified?

p.658, l.7: Where (altitude, latitude) is the majority of Bry in the form of Br and BrO?

p.659, l.9: In the TTL the local lifetime of CH2Br2 is even much longer than 120 days (Hossaini et al., 2012)

p.659, l.12: Note the different definitions of "Western Pacific" in this study and other studies, which may explain some of the differences in seasonality between the studies.

p.660, l.7: Is there active washout above the tropopause in the model? I'm surprised that there is "in-cloud rainout" in the mid-latitude lower stratosphere - or did I misunderstand this statement? Please provide more details on the process.

p.660, l.19: Why are surface concentrations higher in the Indian Ocean in this model simulation? The following paragraph remains somewhat unspecific.

p.661, l.6: 0.88 ppt of CHBr3 (which would be a lot) or 0.88pt of bromine due to CHBr3?

p.662, l.5: I fully agree with these statements. However, two aspects remain puzzling: How can Bry be transported into the upper troposphere when assumed to be present in the form of the two highly soluble species HBr and HOBr? And secondly, observations suggest that there could be a significant amount of BrO in the troposphere, which would offer a pathway for TST PGI, but is not accounted for in this study.

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p.662, l.18: Just as a side comment, could the relation between convective strength in the tropics and large-scale descent in mid-latitudes be used to better constrain convective uplift? I guess it should be possible to validate large-scale descent from temperature and trace-gas observations better than the convective processes.

p.662, l.18: You may cite Aschmann et al. (2011) here, who found a qualitatively similar behaviour for changes of SGI and PGI under El Nino versus La Nina conditions: While SGI slightly increased under enhanced deep convection, the sum of SGI and PGI decreased.

p.664, l.3: Again, it is critical that you specify clearly if full chemistry was used here also in the TTL, or only above the cold point.

Technical corrections:

p.652, l.14: of the VSLS

p.653, l.5: VSLS contribute

p.654, l.24: enter the stratosphere

p.660, l.11: please define LS and MBL when first used

p.661, l.22: that enter

p.664, l.14: “scavenging in” or “scavenging of”?

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 651, 2014.