

***Interactive comment on* “Estimating the contribution of bromoform to stratospheric bromine and its relation to dehydration in the tropical tropopause layer” by B.-M. Sinnhuber and I. Folkins**

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We thank referee #2 for the detailed review and constructive comments.

In our point-by-point response below we show the original comments of referee #2 in italics and our reply in plain text.

Would it be better to assume a constant washout for BrY below 10 km to represent large-scale removal by precipitation, and use a separate parameter for washout between 15.5 km to 17 km to simulate removal by ice?

In our model calculations we have assumed a constant washout rate below 17 km. However, as we do not consider entrainment of bromoform or BrY (the detrainment mixing ratio is a free parameter here) the results above the level of zero radiative heating are independent of the washout rate in the troposphere below. I.e., using different rates for the free troposphere and the TTL may improve the modelled bromoform profiles but will not affect our conclusions. Anyway, as knowledge on washout rate is very limited (depending also on possible heterogeneous bromine recycling, see comment by referee #3) we have decided not to include another parameter for washout below 10 km.

A minor point. It would be less confusing if the authors try to make the distinction between tropospheric concentration and stratospheric concentration. The mixing ratio in equation(1) should really be averages for the troposphere. The tau-chem term in equation (1) refers to the average lifetime below 17 km. It would be better to use a different notation to distinguish that from the tau-chem in equation (2) which is the local photochemical loss as a function of altitude. Finally, the expression on line 25 of p. 12941 is the steady state mixing ratio of BrY in the stratosphere, not the amount of bromine transported to the stratosphere. To get that, one would have to divide by the residence time in the stratosphere.

Okay, in the revised version of our manuscript we have changed the sentences accordingly.

The following will not change the conclusion of the paper. I have some difficulty understanding some of the results. I look upon equation (2) as a 1-D equation for three separate domains: ground to 15.5 km, 15.5 km to 17 km, above 17 km.

(1) It is not clear to me whether 1 pptv was imposed as a fixed mixing ratio boundary condition for CHBr₃ in solving the equation in the first domain. I would like to know if one solves the equation without the bottom fixed mixing ratio boundary condition, what value would one get depending on the imposed flux boundary condition (zero flux, or

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an estimated emission rate to balance the downward advection to obtain 1 pptv). If a fixed mixing ratio boundary condition is used, one has to do a sanity check to see if the flux is reasonable. One can do this by comparing the integrated detrainment source and see whether it balances the photochemical plus detrainment loss. The difference is the artificial source in the boundary layer. to the first domain.

For the model calculations here we have assumed a fixed boundary layer mixing ratio of 1 pptv. We have now included a paragraph on the emission flux needed to maintain the modelled bromoform profile:

The flux of bromoform into the model atmosphere in order to maintain the modelled steady state bromoform profile for a boundary layer values of 1 pptv equals 1.1×10^{12} molec/cm²/day. If one assumes a uniform oceanic source this corresponds to a global flux of 0.6 Tg CHBr₃/year (taking the area of Earth' oceans as 3.61×10^{14} m²). This value is higher than the one given in the previous WMO/UNEP assessment (0.2 Tg/year) but agrees (in particular when scaled to a boundary layer value of 0.75 pptv) with the modelling studies of Nielsen and Douglass (2001) (0.5 Tg/year) and Yang et al. (2005) (0.4 Tg/year).

(2) Above 17 km, there is no washout for BrY, and the detrainment rate is zero. CHBr3 in the stratosphere is maintained by the boundary mixing ratio at 17 km, balanced by local photochemical loss. For BrY, there is no local loss. The question then is how does one reach a steady state condition. If one looks at total bromine, there is no production and removal term. There is an upward advection velocity at 17 km. One can get a steady state with constant mixing ratio only if the advection at the top boundary balances the inflow at 17 km.

Because there is no production or loss of total bromine in the stratosphere (more specifically: above the cold point) in our model the total bromine mixing ratio entering the stratosphere determines the mixing ratio for the whole stratosphere. Although we do not explicitly model this here, it implies indeed that the flux of bromine into the strato-

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sphere is exactly balanced by a flux through the top boundary.

(3) Finally, I wonder if numerical diffusion is affecting the solution.

The upwind scheme used here is in principle numerically diffusive. We have tested the effect of numerical diffusion by performing model calculations with a range of different time steps (which should affect the numerical diffusion) but found no significant changes in the model results. Consequently we believe that numerical diffusion is not a major problem for our results presented here. Further confidence comes from the fact that the model agrees well with observations of ozone and CO.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 12939, 2005.

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