**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "Modeling the transport of very short-lived substances into the tropical upper troposphere and lower stratosphere" *by* J. Aschmann et al.

J. Aschmann et al.

jaschman@iup.physik.uni-bremen.de

Received and published: 21 September 2009

We thank the reviewer for her/his thoughtful comments and suggestions for improvements. In the following, the original remarks of the reviewer are in *italics*.

The most substantive question I have is regarding the effect of freezing on soluable species (Bry degradation products). If there is HBr and HOBr in solution and cloud drops freeze, what happens? Is the Bry not ejected? And the authors hint at this, but is Bry able to attach to or be bound in ice clouds that form in situ (not from convective detrainment).
A detailed description of the micro physics and multiphase chemistry involved is diffi-

cult and beyond the scope of our present study. The scenario we have in mind is not primarily the lifting of HBr and HOBr in cloud drops, because it seems unlikely that this is a significant source of inorganic bromine for the TTL, but rather the uptake of HBr and HOBr (formed from the degradation of VSLS in the TTL) on ice particles. Heterogeneous reactions may activate HBr and HOBr on the ice surface so that reactive bromine (Br or BrO) could be released back into the gas phase. This process is actually included in many stratospheric chemistry models in the context of PSC chemistry and we are currently performing a follow on modeling study were we consider this process for the TTL in more detail.

Pg 1, Intro, para3: When discussing the level of zero radiative heating, you might want to give a reference. E.g.: Gettelman et al 2004 or Corti et al 2005
Pg2, 2nd para: A sentence or two on the other bromine models (Kerkweg, Gettelman) would be appropriate here.
pg5, section 3.2: regions of ttl dehydration: a better reference than Holton and Gettelman would be Gettelman et al 2002b (which does show regions).
We will include the suggested references.

Pg2. Sec 2.1, 3rd para: Why did you pick 20 days for Bromoform, not 26?
Based on our previous calculations (Sinnhuber and Folkins, 2006, Fig. 3) we believe that a value of 20 days may be more representative for the TTL than the often used value of 26 days, which is a global average at 5 km altitude (WMO, 2006, Table 2-1). We have also conducted sensitivity runs where we set the lifetime of our idealized bromoform tracer to 26 days. This changes calculated tracer mixing ratios slightly but does not change our main conclusions.

Pg3, equation 1: I must have missed something, but the units of eq 1 seem strange: f (tracer flux) has units of kg-tracer/s ?
*then f= dc (kg-air/s) * m (kg-air) * [X] kg-tracer/kg-air*
Seems to me the m term is not necessary here. What did I miss?
We realize that our description of the formula is not complete. The original ECMWF

detrainment rate has the units kg/(m3 3h) (the 3 hours come from the forecast step). We integrate the detrainment rate over the corresponding grid box and divide it by the box density, so our resulting dc has the units 1/s. We will clarify our description of the formula accordingly.

Pg3, 2.3, water vapor. The relaxation to a maximum seems strange, and seems a bit pathological (maybe), I assume you will not have a H2O sink if H2O greater than 6.8 ppmv? Why did you not just use something like dH2o/dt = 2[CH4]/tau?*
We don't have CH4 in our model so we can't take methane oxidation explicitly into account. While in principle this could be an H2O sink if H2O is higher than 6.8 ppmv and less than the saturation mixing ratio, in practice this does not play a role since the lifetime tau is 6000 days at 100 hPa.

Pg4, end of sect 2: Does methyl iodide work the same way?
The degradation of methyl iodide is handled the same way as for bromoform, besides the shorter lifetime of only 5 days. The main difference is that methyl iodide is just removed from the model whereas bromoform degrades into Bry.

Pg 4, sect 3.1.1: "There is in general good agreement..." please make good quantitative. Can you put the model standard deviation for example on the plots? This would help (you could test for the difference of means, or state they are not statisitically different).
This is a good point. We will add the model standard deviation to an updated version of this plot.

Pg4, sect 3.1.1: "notable differences" : what are they?
The idealized CHBr3 tracer profile agrees with the Pre-AVE measurements, however, compared to the Cr-AVE data it shows in general to high concentrations. As we have stated, that might be due to inaccuracy of the model or simply due to the fact, that the time and area of the Cr-AVE campaign is different from the Pre-AVE campaign.

Pg4, sect 3.1.1": "high bias in the observations" relative to what? It seems dangerous

to assume your model is unbiased. Mostly this is a semantic argument, unless you have reasons to believe there are problems with the measurements for this campaign. If so, please state them. It might mean that there is VSL well above the tropopause and the model is wrong.
There are indications (in particular for the CH3I measurements during Cr-AVE) for a small high bias in the observations due to a possible residual artifact observed in new canisters that were used during Cr-AVE. We will include a statement in the manuscript.

Pg4, section 3.1.2: can you separate the differences between convective transport and chemistry with regard to ozone? This could be critical for evaluation of the model. What about biases in transport outside of convection?
The important point is, that the convective source [O3]c is uniform and also the ozone production/loss due to the linearized O3 chemistry as well as the initial ozone tracer setup is identical for these two stations. Since the modeled profiles are able to reproduce the sonde measurements in general the differences must be due to the convective transport in these areas. Whether the deviations from the observations are originated from the convection or ozone production one cannot decide. Concerning the transport outside of convection: Of course the large-scale transport can be biased too but we believe (a) the large scale transport constrained by diabatic heating rates is more robust than the convective transport and (b) the bias is larger in the convectively more active regions, indicating a relation to the convective transport.

pg 6, end of section 3: Careful. You have earlier said convection is not necessary up to the top of the TTL. Also you might want some references for the regions where convection is deepest: Try Liu and Zipser 2005 or Gettelman et al 2002c
Thanks for pointing to this apparent contradiction. At the end of section 3.2 we stated that convection **above** 380 K doesn't seem to be important for our results relating to stratospheric bromine loading. The convective activity between level of zero radiative heating and the coldpoint (where the description "high-reaching" is justified) is crucial for the uptake of bromine. We will modify the text accordingly to make this point clear

and include the suggested references.

Conclusions: There are now several estimates from simulations of the estimated quantity of VSL bromine into the stratosphere. Can you summarize these results in the context of that other work. Does it agree or not? I think the answer is that it does agree. Should the earlier estimates be increased due to some soluable species?

We agree that it would be appropriate to put our results into context of earlier estimations. Considering the uptake of bromoform our results do indeed agree with recent studies taking into account the differences in the modeling approaches, source strengths, lifetimes and so on: Tropical mean bromoform at 18 km altitude:

- Warwick et al. (2006): 0.1 ppt

- Hossaini et al. (2009): 0.15 ppt

- Gettelman et al. (2009): 0.5 ppt

- our results: 0.1 ppt

Comparing the estimations for bromine loading due to VSLS is more difficult because of some significant differences between the conducted studies. Warwick et al. (2006) give an estimation of 6 to 7 ppt of additional inorganic bromine due to VSL bromocarbons at about 18 km (for bromoform alone about 3.4 to 4 ppt). Gettelman et al. (2009) give a range of 1.1 to 4.1 ppt (bromoform only: 0.3 to 2.1 ppt) for the same quantity. Our estimations of additional bromine loading due to bromoform (1.6 to 3 ppt) are in between.

We will add an additional paragraph discussing these results into our conclusion section.

References:

C5063

Gettelman, A., Lauritzen, P. H., Park, M., and Kay, J. E., Processes regulating short-lived species in the tropical tropopause layer, J. Geophys. Res., 114, D13303, doi:10.1029/2009JD011785, 2009.

Hossaini, R., Chipperfield, M. P., Monge-Sanz, B. M., Richards, N. A. D., Atlas, E. and Blake, D. R., Bromoform and dibromomethane in the tropics: a 3-D model study of chemistry and transport, Atmos. Chem. Phys. Discuss., 9, 16811-16851, 2009

Sinnhuber, B.-M. and Folkins, I., Estimating the contribution of bromoform to stratospheric bromine and its relation to dehydration in the tropical tropopause layer, Atmos. Chem. Phys., 6, 4755–4761, 2006.

Warwick, N. J., Pyle, J. A., Carver, G. D., Yang, X., Savage, N. H., O'Connor, F. M., and Cox, R. A., Global modeling of biogenic bromocarbons, J. Geophys. Res., 111, D24305, doi:10.1029/2006JD007264, 2006.

WMO (World Meteorological Organization), Scientific Assessment of Ozone Depletion: 2006, Global Ozone Research and Monitoring Project-Report No. 50, 572 pp., Geneva, Switzerland, 2007.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 18511, 2009.