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Comment

## ***Interactive comment on*** “Impact of deep convection and dehydration on bromine loading in the upper troposphere and lower stratosphere” *by* J. Aschmann et al.

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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*.

*1. Section 2.2, 3rd paragraph. My understanding is that the soluble and insoluble Bry are two parallel tracers, representing the two extreme cases to examine the upper and lower bound of bromine loading. However, later in section 3.2.1, 2nd paragraph, the authors mentioned “another aspect that influences our results of the idealized setup is the assumption regarding the partitioning of soluble and insoluble*

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Bry". From what described in section 2.2, there seems to be no partitioning between the soluble and insoluble Bry. In both extreme cases, Bry exists either all in soluble or insoluble form. Please clarify.

Your understanding, that soluble and insoluble Bry are two parallel tracers is correct. We agree that our phrasing in the second paragraph of section 3.2.1 is mistakable. The "partitioning" of soluble and insoluble Bry relates to our assumption that the soluble Bry tracer is uniformly soluble, which overestimates the washout in polar regions. Therefore the sensitivity calculation restricts the washout between 40N-40S, taking into account that the fraction of HBr to Bry is small at higher latitudes. We will change the paragraph accordingly.

2. Section 2.3, the part on heterogeneous chemistry. I feel this part needs some clarification or reorganization for a clear explanation of how heterogeneous chemistry is handled within the model. First, the authors mention that SLIMCAT incorporates an explicit treatment of uptake of halogenated species on liquid particles, but the description is vague. It would be helpful to add a word or two explaining how it is done and for which species (HBr? HOBr? BrONO<sub>2</sub>?) In the sensitivity simulation (shown later) with heterogeneous activation turned-off, does it only refer to the reaction on ice particles or on both ice and liquid particles? I might be wrong on this, but from the text, it seems that the authors are only concerned about the heterogeneous chemistry on ice particles. Do we have any knowledge from previous published literature of the relative importance of solid vs. liquid phase heterogeneous bromine chemistry, particularly in the UT/LS? If so, please add the references. Secondly, the sentence "After one model timestep all dissolved and adsorbed species are released back into gas phase instantaneously" on page 8 (line 6-7) is very confusing. How does the model handle the physical and chemical processes that relates to heterogeneous chemistry? I would guess the sequence is dissolving/adsorption - sedimentation and heterogeneous chemistry - release back to gas phase, right? It makes sense to release the insoluble species (e.g. Br) back to the gas phase after each time step. But

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for soluble and adsorbed species (e.g. HBr), shouldn't they remain within the ice/liquid particles and convert back to gas phase only when evaporation happens?

The mechanism of uptake of halogenated species on liquid particles is detailed in Chipperfield (1999) and was not changed for this study. Therefore, our description concentrates on the adsorption process on ice particle surfaces which was developed in the course of this work. However, we agree that a short overview of the other uptake processes would be helpful. It will be included in the revised manuscript.

Regarding the sensitivity run without heterogeneous activation: All heterogeneous reactions listed in Table 2 are switched off. Those reactions occur on NAT and ice particles as well as on liquid aerosols. However, the given gamma values in Table 2 only apply to the reactions on ice surfaces. We will clarify this in the upcoming revision.

Finally, the model does not contain an explicit treatment of evaporation. At the beginning of a chemistry time step, trace gases can be dissolved or adsorbed on particle/aerosol surfaces if thermodynamically possible. Then the chemical integration is calculated including the heterogeneous reactions. After that all dissolved and adsorbed species are released back into gas phase instantaneously. We will add a sentence to clarify this issue.

*3. Page 10 line 19-page 11 line 3 and figures 4 and 5. I think Figure 4 is a very informative figure that helps to explain the difference between the idealized setup and full chemistry scheme. Figure 4 suggests that only in the tropical UT/LS, Bry is mainly in the soluble form. Therefore, the impact of scavenging is much smaller in the full chemistry setup than in the soluble Bry case in idealized setup. However, this discussion probably fits better in section 3.2.2. On the other hand, what the authors try to explain with figure 5 is not that important. The idealized setups with soluble and insoluble Bry are just two extreme cases helping to illustrate the upper and lower bound. Assuming everything is in soluble form might not be appropriate for the south pole, but it is just a trivial detail since the idealized setup is not meant to represent the*

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real atmosphere. I would suggest: i) delete Figure 5, ii) move Figure 4 to section 3.2.2 and elaborate more on why the full chemistry differs from the idealized setup in the contribution of VSLS to stratospheric bromine.

We agree that Figure 5 can be discarded since it covers only a side aspect of our study. We will rearrange the affected sections.

4. Page 15, line 6-9. Unfortunately, I don't find the spatial pattern of total bromine very similar to that of the RH. In general, convection affects total  $B_{ry}$  in two ways: i) increase  $B_{ry}$  through injection of source gas, ii) decrease  $B_{ry}$  through scavenging of product gas (tied to changes in RH). The pattern of total  $B_{ry}$  in the Pacific is regulated by both factors, while the negative anomaly over the Indian Ocean is dominated by the decrease in source gas injection (as you can see in TT20 and TT120). Similarly, during the La Nina year, total  $B_{ry}$  is also affected by both, with the increase due to decrease in scavenging more apparent in the northern tropics and the decrease due to decrease in source gas injection more apparent in the southern tropics.

We agree that our description on page 135 is not precise enough and may lead to misconceptions (see also reply to point 5 of the referee's comment). Total Br cannot be altered by convection alone in our model setup, because the increase in source gases is completely balanced by the dilution of  $B_{ry}$  (detrainment mixing ratio of  $B_{ry} = 0$ , see p. 134, l. 4 et seq.). However, since the soluble  $B_{ry}$  tracer is affected by washout there can be variations in total Br, but they are solely related to washout. Therefore, the main features of the total Br anomalies during El Nino 97/98 depicted in Fig. 10, the positive equatorial anomaly in the Pacific neighbored by the negative anomaly at around 20N/S, can be directly related to the RH distribution. We will try to make this more clear in the revised manuscript.

5. Page 15, line 9-13. From Figure 10, the spatial distribution of TT20, TT120, and hence total  $B_{ry}$ , during the La Nina conditions do not simply look like the opposite of the El Nino conditions. In particular, TT 20 and TT120 show a positive anomaly in

*the central and eastern Pacific but a negative anomaly near the western Pacific. It worth to spend a bit more effort in explaining how the atmospheric circulation change from El Nino to La Nina years and how this impact the vertical transport of source gases and soluble product gases.*

We concur with the reviewer that the paragraph on page 135 needs some clarification as stated in the comment to the previous point. Whereas the main features of the SST, TT20 and TT120 anomaly patterns during the La Nina season are roughly similar to those of the El Nino 97/98 (with opposing sign) there is no comparable dipole-type structure in the detrainment rate and relative humidity. Consequently, the anomalies of total Br look different. The positive anomaly in the Northern Hemisphere can be explained with the low relative humidity in this region which likely restricts washout of soluble Bry. However, this explanation is not applicable for the negative anomaly in the Southern Hemisphere. It is possible that this anomaly is caused by transport of Bry-depleted air from India/northern part of the Maritime Continent region, where a positive anomaly of RH exists. We will add a corresponding discussion in the revision.

*6. Finally, this paper presents a range of estimates of the contribution of brominated VSLS to stratospheric bromine under varying assumptions and model setup. I think it would be very useful to add a table summarizing the estimated contribution from all the experiments mentioned in this study. This would make it easy for the readers to comprehend and remember the relative importance of individual source gas (e.g. CHBr3 vs. CH2Br2) and different processes (heterogeneous chemistry, soluble vs. insoluble assumption, etc.)*

That is a very good idea we will consider for the updated manuscript.

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