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

Anonymous Referee #1

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This paper presents a comprehensive modeling analysis of the impact of dehydration and deep convection on the amount of stratospheric bromine. It provides a quantitative estimate of the contribution of the two most important brominated short-lived substances, CHBr3 and CH2Br2, to stratospheric bromine under different model set up (idealized setup with varying dehydration assumption as well as full chemistry simulation). It also addresses an important question – how will this contribution vary during different phases of ENSO, which has not been discussed in previous literature. The experiments are thoughtfully designed and well executed. I recommend the paper be published in ACP after addressing the following comments.

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 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.

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? BrONO2?) 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 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?

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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 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.

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 Bry in two ways: i) increase Bry through injection of source gas, ii) decrease Bry through scavenging of product gas (tied to changes in RH). The pattern of total Bry 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 Bry 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.

5. Page 15, line 9-13. From Figure 10, the spatial distribution of TT20, TT120, and hence total Bry, 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.

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.)

Minor comments:

1. Page 6, line 5: CHBr3 and CH2Br2 were introduced in section 1 already.

2. Page 13, line 17-18: Change "almost all detrainment and also dehydration occurs . . ." to "almost all detrainment and dehydration occur . . .".

3. Page 15, line 5: a "," is missing after "relative humidity".

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 121, 2011.