

Hamer et al. (2013) presents a modeling case study together with field measurements obtained during the SHIVA campaign to analyze transport and chemistry of CHBr₃ and its product gases within deep convective systems. In addition to this, the authors also aim to identify how results from the episodic analysis relate to those learned from CTM and CCM framework. This work is interesting since it is one of a few model studies, if not the only one, that examine what happens inside episodic convective lofting, in contrast to the majority of previous published work focusing on simulation of bromoform chemistry and transport in a climatological perspective. However, after carefully read through the paper, I feel in the current form, there are significant issues remain with the overall model performance which affect the soundness of the conclusions. I would recommend major revision is needed before the paper be considered for publication in ACP.

Major concerns:

1. Based on the results presented in this version of the manuscript, the model's performance in a quality representation of CHBr₃ and the related photochemistry are questionable. Here are the specifics:

- i) At several places in the manuscript, the authors state that the model performs well in simulating CHBr₃ in the marine boundary layer and in the convective outflow. I have to say I do not agree with this statement. The comparisons in Figure 13 and Table 4 show that modeled CHBr₃ is about 2-3 ppt near the surface, compared to the observed ~1 ppt level (a factor of 2-3). In the upper troposphere, the modeled value is about 1 ppt, while the observed mixing ratios vary from 0.5 to 1 ppt. None of the model series shown in figure 13 falls within the instrument uncertainty range of the observations. Secondly, a simple calculation suggests that the ratio of the UT and BL values also differ quite a bit between the model and the observations, implying biases in the simulated lofting efficiency. Here, I would suggest the authors at least add one figure showing the mean vertical CHBr₃ profile (altitude vs. concentration), which will clearly illustrate both issues. Thirdly, 4 of the 5 panels in Figure 13 suggest the model also fails to capture the observed variability of CHBr₃. In panel d, the simulated CHBr₃ is actually anticorrelated with the observed time series. Within a regional modeling framework, which mimics the real meteorology, this likely implies issues with surface emission and concentration.
- ii) The second issue I have is the inadequate evaluation and discussion of model OH within the convection column and outflow (p20634). The authors stated that "We perform a comparison between the model's simulated OH and the various reported climatologies and observations ... The model simulates very similar vertical profiles ... and we therefore conclude that the quality of the OH simulation is sufficient". OH abundance involves a complex fast photochemistry and is determined by a delicate balance between its sources and sinks. Within convectively lofted pollution plumes, unlike the background atmosphere, the OH photochemistry becomes particularly complex, e.g. in addition to O₃ photolysis, HCHO and acetone can act as additional source gases of OH and wet removal of H₂O₂ and HNO₃ provides additional loss pathways. As a result, OH level within convective pollution plume can deviate significantly from its climatological mean and vary by orders of magnitude. Since reaction with OH is the main loss

pathway of CHBr₃ and OH can increase significantly within convection plumes therefore shorten CHBr₃ lifetime from a few weeks to several days, the model's ability to reproduce OH within convection plumes becomes a critical issue and comparing OH with climatological values appear to be inadequate in this case. While observational OH may not be available for model validation in this case, since the model does a poor job in simulating important source and sink gases for OH such as O₃ and CO (see figure 14), implying there are likely issues with simulated OH.

- iii) In section 4.1.4, the authors discussed the existing issues within the model in overestimate in rainfall rate up to a factor of 5, implying excessive washout of soluble product gases.

The combination of the above three problems suggest there are major issues with CHBr₃ chemistry, abundance, vertical transport efficiency and washout rate of its soluble product gases, all are critical in CHBr₃ chemistry modeling. These significantly impair the robustness of the conclusions of how much CHBr₃ can be efficiently transported from the marine boundary layer to the upper troposphere and the relative contribution of source gas injection vs. product gas injection.

2. One main objective of this work is to “identify how results from the episodic analysis relate to those learned from CTM and CCM framework”. To address this, the authors have referenced in several places how their results are compared to previous work, but the discussion remain vague. One particular issue I had is that most of the previous work referenced are modeling studies based on annually averaged global mean results and focus on the entrance into the lower stratosphere (above ~12-14 km in the tropics), which reflects transport and chemical processes on a very different temporal scale and likely involve other essential dynamical processes, e.g. radiative ascent, TTL venting into the lower stratosphere (LS). This work only presents results from a 3-day model simulation in a couple of episodic convective events that likely only reach the upper troposphere, or at most the base of TTL. The outcome, e.g. the amount of remaining CHBr₃ and SG and PG ratios can be very different in these convective outflow regions than those values from the lower stratosphere as presented in many of the previous CTM and CCM studies. An in-depth discussion on how these results can be linked to CTM and CCM framework is still lacking, in particular the essential piece on how to reconcile the differences in the spatial and temporal scale between a regional model that focus on episodes and the global scale CTMs and CCMs is yet absent.

3. In the introduction, the authors mentioned the importance of HCHO+Br and BrO+NO₂ reactions in bromine simulation. While a short discussion on elevated HBr and BrONO₂ within pollution plumes at 3km when HCHO and NO₂ are elevated is presented (p20638-20639), it seems to me the authors stop-short in explaining how the addition of above reactions affects CHBr₃ product gases in the upper troposphere convective outflow, which are the key focus of this paper. Also the discussion in this section was not well organized and hard to follow. It would be useful to quantify the importance of these two reactions as the significance of these on troposphere-to-stratosphere transport of bromocarbons will be appealing to the modeling community.

Minor comments:

1. P20616, L24-25: You may want to add reference to Aschmann and Sinnhuber (2013), which also speciates inorganic Br₂ into soluble and insoluble forms.

Aschmann, J., and B.-M. Sinnhuber (2013), Contribution of very short-lived substances to stratospheric bromine loading: uncertainties and constraints, Atmos. Chem. Phys., 13, 1203-1219, doi:10.5194/acp-13-1203-2013.

2. Section 2 seems to be somewhat out of place and interferes with the flow of the paper. It might fit better at the beginning of section 4.

3. P20621, L16-30. The description is too general. Even when all the chemical reactions are included in the Appendix, it will be nice to add a few sentences here to describe the chemical scheme, e.g. how many chemical species, how many reactions, is it mainly stratospheric chemistry based or tropospheric chemistry based or combined, etc.

4. P20622, L15: Add “,” after “its transport”.

5. P20626, L12: Add “,” after “qualitatively well”.

6. P20626, L17: Change “time” to “temporal”.

7. Personally I feel section 4.1 is a bit too long and can be condensed a bit, since the paper is already very long and this part is relatively less essential compared to the rest.

8. Table 1. Aschmann et al. 2010 should be Aschmann et al. 2011. Also both Aschmann et al. 2009 and 2011 set constant VMR at the base of tropopause, not boundary layer.