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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Bromine from short–lived source gases in the Northern Hemisphere UTLS" *by* Timo Keber et al.

Anonymous Referee #3

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The manuscript by Keber et al. describes measurements of short-lived organic bromine gases from several airborne campaigns in the UTLS mostly north of 40° latitude in different seasons. The authors then evaluate how different combinations of models and emission scenarios compare to observations. The study is relevant to better quantifying the role of organic bromine in catalytic cycles that deplete stratospheric ozone. The main points of the paper are the demonstration of a latitudinal gradient in UTLS organic bromine, a winter maximum in VSL Br gases in the UTLS, recognition that VSL Br concentration near the extratropical tropopause is greater than that at the tropical tropopause, and that current emission scenarios show a range of imperfections that limit their accuracy. I might argue that all of these conclusions are not new, but the measurements, importantly, quantify the levels of UTLS organic Br that provide a

solid basis for more detailed and quantitative analysis. Thus, the manuscript will be a valuable contribution to the further understanding of halogen chemistry in the lower stratosphere.

I think the strength of the manuscript is in the presentation of the data and identification of the seasonal and latitudinal variability in the measured Br gases. However, much of the paper focused on the model and emission scenario comparisons to the data. The authors state in their summary, "While it is not the main purpose of this paper to evaluate emission scenarios. . ." they nonetheless spend a large amount of effort to do just that, and they do so in more detail than is needed to demonstrate their point that models have problems. If model comparison to data is a significant part of the message, then I would like to see (perhaps in the Supplement) how the models compare to measurements for CFCs, halons, CH4 or other gases with better understood tropospheric distributions. Does model SF6 compare well to the binned measurements from these missions? This would help provide some context for looking at the Br distributions and their deviations from model predictions. My suggestion, though, is not to add more model discussion in this paper, but to reduce the level of different comparisons to models that is done in the text and just highlight major issues associated with emission scenarios.

I also had some trouble sorting through the different manipulations that the authors used to help with the comparisons between measurements and models. The authors go into great detail in how they adjusted and binned the data to presumably reduce variability from variations in dynamics and transport, and to provide a more robust comparisons to model outputs. While I would agree that these might be reasonable adjustments, I found the multiple presentations confusing and left me wondering about what I was viewing in the plots and how the data might look with no adjustments. Perhaps the authors can think about possibly simplifying (or better explaining) the discussion of the adjustments. Even though the goal of the adjustments was to reduce variability due to dynamical factors, the resulting distributions retained significant spread in the data,

particularly within -+40K of the tropopause. The authors chose to focus on the average profiles, but it would seem that a more detailed evaluation of the variation (and its comparison to variance in a model) could produce interesting results on sources, transport and variability of Br in the critical region near the tropopause. Maybe a subsequent paper can do this.

In section 5, I did not understand the need to go through the linear mixing model to show how the emission scenarios that placed high (or low) CH2Br2 (or CHBr3) in the tropical (or extratropical) tropopause were different from measured values. Data summarized in Table 5 is sufficient to demonstrate the impact of "incorrect" modeled organic Br in the tropics or extratropics. Why go through assumptions that aren't necessarily realistic? Or you might be able to improve the mixing model with data from other gases, such as SF6, which can be used to perhaps better estimate tropical vs extratropical fractions of air.

Given the goal to provide a more robust assessment of the role of Br in the lowermost stratosphere at N latitudes, and given the availability of multiple models, I would have found valuable some discussion of the actual impact of different Br levels on the ozone budget of the impacted region. How does an uncertainty of -+ 1 ppt VSL Br propagate to modify ozone destruction rates?

Other comments:

Minor VSL Br. A) I would suggest adding profiles of the minor VSL Br in a Supplement. B) The authors state that "the observed decrease with altitude in the stratosphere is consistent with the relative lifetimes of the different compounds". While generally true, this does not seem to be the case for CHBrCl2 compared to CH2Br2 (page 7 line 15). Lifetimes of these compounds differ by a factor of about 3.5, but the gradients appear close to the same. Perhaps something is wrong with the lifetime estimate? C) Tables 1 and 3 show the mixing ratio of CH2BrCl as 0.1 – 0.2 ppt (100 – 200 ppq), but the GhOST-MS is reported in Table 1 to have a detection limit of 130 ppq (0.13 ppt). Not

sure how this is possible. I note also that the GhOST-MS characteristics are quite different here compared to that reported in Sala et al. (2014). Was there a significant modification to the instrument?

Uncertainties. Page 6, top. Please report uncertainties associated with the total organic bromine levels reported here and elsewhere.

Organic/Total Br. . .I second the recommendation of one of the other reviewers to be more careful in terminology of total bromine/total organic bromine, etc.

Data availability. I did not see that these data are available in any public archive. Please list how the data from these flights can be accessed.

Typo: Table 1 "Reproducibility"

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