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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Finding the missing stratospheric Br_y: a global modeling study of $CHBr_3$ and CH_2Br_2" *by* Q. Liang et al.

Anonymous Referee #1

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The manuscript by Liang and coworkers presents a top down estimate of oceanic emissions of brominated very short-lived substances (VSLS), in particular CHBr3 and CH2Br2, using a global chemistry climate model constrained by an extensive set of aircraft observations. This top-down estimate is consistent with previously published bottom-up estimates and results in an additional stratospheric bromine loading of about 5pptv, consistent with observations of inorganic bromine in the stratosphere. Overall from the present manuscript in line with previous publications there now appears to emerge a consistent picture of the contribution of VSLS to the stratospheric bromine loading for the present day atmosphere. The paper is well written and an important contribution to the current discussion on the role of VSLS for stratospheric bromine.

I recommend publication in Atmos. Chem. Phys. after consideration of the following comments.

One of the strengths of this paper is the inclusion of a relatively large set of aircraft observations to constrain the emission estimate. In this respect I feel the construction of the emission scenario A should be described and discussed in greater detail. It was not clear to me how and why the emission scenario was constructed in this particular way. I assume that some kind of optimization process was involved? Where only the overall emission strengths in the different regions derived, or also the borders of the different regional components varied? Is it possible to provide an estimate of how robust the numbers derived are (e.g., how robust are the 60% from open ocean against 40% from coastal emissions)? What was the rationale behind choosing these particular regional components?

Specific comments:

p. 23625, l. 23: As I will discuss in a bit more detail later on, I was surprised by your conclusions that scavenging of Bry in convective updrafts apparently has so little effect on stratospheric bromine. However, in any case I believe that the statement "Bry_VSLS in the stratosphere is not sensitive to convection" at the end of the abstract should be made more specific to avoid any confusion here.

p. 23626-23627, Introduction: I suggest to include references to the more recent studies by Kerkweg et al. (2008), Gettelman et al. (2009) and Aschmann et al. (2009). In particular as these studies have considered more detailed washout or scavenging than the simple 10-day washout lifetime mentioned (p. 23627, l. 28).

p. 23627, l.3: Is it really justified to say "»100pptv" (which I understand as "much larger than 100pptv"), or would it be more appropriate just to refer to ">100pptv"?

p. 23629, l. 24: Why are there quotes around "transported as an individual tracer"?

p. 23632, l. 20: How strong is the evidence that the emission of CH2Br2 should

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(always?) be proportional to the emission of CHBr3? Can you give a reference here?

p. 23633, l. 5: I don't understand why your model underestimates observations in the marine boundary layer near coast lines. I thought the emissions were derived as to agree with the observations? A few more words here would be helpful.

p. 23634, l. 25: It may be good to discuss (or speculate) in a bit more detail why the strong emissions are confined to 10° N/S. I assume this is related to the oceanic upwelling along the equator? However, if this is true, is 10° N/S really the optimum, or could it also be confined to an even more narrow region?

p. 23635, l. 13: "... difficult to reproduce...": Why is this difficult to reproduce in a top-down estimate?

p. 23636, l. 18: It is a long shot to imply from the STRAT observations that CHBr3 has increased in the lower stratosphere and that this increase is a result of changes in chemical loss and/or troposphere-to-stratosphere transport. As chemical loss of CHB3r is dominated by photolysis, how/why should this have changed since the mid-1990s? Changes in transport are in principle more plausible; however, this is an important issue here. Do you have any indications from the model for large inter-annual changes of troposphere-to-stratosphere transport of CHBr3? Not only do surface observations of CHBr3 not show any clear large inter-annual changes, but also there is little evidence from stratospheric observations of inorganic bromine for significant inter-annual variability (e.g., WMO, 2007). Stratospheric observations reported by Sturges et al. (2000) are predominantly from northern hemisphere mid- and high latitudes, so I would not really expect to find high CHBr3 mixing ratios in these observations. Which 1997 measurements are you specifically referring to? I do agree that it may not be possible to finally solve the discrepancy between model and observations for the STRAT campaign. However, I don't agree that this question is "beyond the scope of this paper" (p.23637, l.3).

p. 23637, Fig. 10: Could you comment on the differences between model and ob-

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servations of CH2Br2 in the lower stratosphere between 100 and 50 hPa seen in Fig. 10?

p. 23639, l. 19: A few more words on the possible reasons and implications for the large uncertainty could be helpful.

p. 23640, l. 14: "seen" -> "see"

p. 23641, l. 14 and end of Conclusions section: I am surprised to see that scavenging in convective updrafts apparently has only a very minor impact on the Bry from VSLS delivered into the stratosphere. In particular as you have assumed (a) that all inorganic bromine will be highly soluble and (b) Bry is removed completely when convective updrafts are encountered. From these two assumptions I would have assumed that you possibly overestimate the sensitivity to scavenging in the model but instead you find only a minor effect. Why is this so? What does this imply? Does that mean a large fraction of the troposphere-to-stratosphere transport takes place outside of convective updrafts? A bit more discussion would be good here, in particular as this is "contrary to the conventional wisdom" (p. 23641, l. 15). I feel the reference to Hossiani et al. (2009) is slightly out of place here as they don't explicitely consider scavenging.

Finally, I suggest that you discuss briefly how your results compare to other published results on the amount of VSLS delivered into the stratosphere and the relative contributions of source gas and product gas injection.

References:

Aschmann, J., Sinnhuber, B.-M., Atlas, E. L., and Schauffler, S. M.: Modeling the transport of very short-lived substances into the tropical upper troposphere and lower stratosphere, Atmos. Chem. Phys., 9, 9237-9247, 2009.

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.

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Kerkweg, A., Jockel, P., Warwick, N., Gebhardt, S., Brenninkmeijer, C. A. M., and Lelieveld, J.: Consistent simulation of bromine chemistry from the marine boundary layer to the stratosphere Part 2: Bromocarbons, Atmos. Chem. Phys., 8, 5919-5939, 2008.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 23623, 2009.

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