

We thank the reviewer for the thoughtful and useful comments. Our responses to the comments are provided below, with the reviewer's comments italicized.

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?

We derive the emission distribution of CHBr_3 following a similar approach to that introduced in Warwick et al. (2006) and use Scenario 5 of Warwick et al. (2006) (Scenario C below) as our baseline emission. We use 10 individual components to represent emissions from the open ocean and coastlines near the equatorial tropics, and at the mid and high latitudes, respectively. Multiple sensitivity runs were conducted by varying the magnitude of emissions from each region and the latitudinal border of each region. The optimum emission estimate for each region is achieved by simultaneously matching i) the observed concentration in the middle troposphere and ii) the observed vertical gradient in the corresponding region. For clarity, we present in this study only the optimum emission estimate (Scenario A). Due to the simplicity of the approach (as compared to inverse modeling studies that include a more complicated mathematical calculation), it is not possible to provide a quantitative estimate of the uncertainties in our emission distribution. Our sensitivity simulations show that vertical gradients of CHBr_3 in the coastal regions are sensitive to the magnitude of coastal emissions. However, as we have discussed in section 3, due to the model's inadequacy in capturing the strong localized emissions along coasts, the 40% from coastal emissions (60% from open ocean) is very likely an underestimate. We have added this discussion to the manuscript.

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.

We have changed it to "Our sensitivity study with no wet scavenging in convective updrafts suggests that amount of Bry from VSLS in the stratosphere is not sensitive to convection. Convective scavenging only accounts for ~0.2 pptv (4%) difference in inorganic bromine delivered to the stratosphere." to make the statement more specific.

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).

We thank the reviewer in pointing out the importance of the above studies, which we have referenced in our revised manuscript where appropriate.

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"?

We have changed it to “>100 pptv”.

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

The quotes were added due to a mistake during format editing. We have deleted the quotes in the revised manuscript. We apologize for the confusion that may have caused.

p. 23632, l. 20: How strong is the evidence that the emission of CH₂Br₂ should (always?) be proportional to the emission of CHBr₃? Can you give a reference here?

Measured concentrations of CHBr₃ and CH₂Br₂ in the coastal marine boundary layer are highly correlated (Yokouchi et al., 2005), suggesting they are produced by similar marine macroalgae sources and therefore have similar source distributions (Carpenter et al., 1999). Atmospheric measurements from the aircraft campaigns used in this study also show consistently high correlations (correlation coefficients range between 0.59 and 0.96 for individual missions) between the measured concentrations of CHBr₃ and CH₂Br₂ below 1 km altitude. Therefore, it is reasonable to employ the same distribution for CH₂Br₂ as that derived for CHBr₃. We have added this to the text.

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

In general, top-down estimates derived using global chemistry models rely on background concentration in the free troposphere. As a result, the derived estimates likely underestimate the global source as they miss the influence of strongly localized sources (with concentrations much higher than background concentrations), e.g. along coasts and edges of ice sheets. These localized sources are confined to regions much smaller than a typical model grid box (2° latitude by 2.5° longitude in this study, ~200km×250km). If the model were to fill the entire grid box with emissions that yield the observed localized high concentrations, the simulated concentrations in the background atmosphere (further away from the coastal source regions) will be too high compared with observations. Therefore, even with a top-down emission estimate, it is difficult to reproduce the observed high concentrations near the surface while matching the background observations in the troposphere at the same time. We have clarified this in the text.

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?

High emission near the equator (between 10°S-10°N) is likely associated with tropical upwelling and active planktonic production, as previous observed in the equatorial Pacific (Atlas et al., 1993) and the tropical East Atlantic Ocean (Class et al., 1986). Based on the observations used in this study, confining strong emissions between 10°S-10°N is optimal. Narrowing the high

emission band to 5°S-5°N, on the other hand, would have led to a simulated band of high concentration that is too narrow when compared to the observations at 2.5°×2° resolution.

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 CHBr₃ 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 interannual 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).

We greatly appreciate the reviewer's insightful comments. Based on comments from the reviewer, we have examined in more detail the possible reasons why the STRAT measurements show very low CHBr₃ concentrations compared to the other missions. There are two possible reasons. First the difference was possibly due to the natural variability of short-lived species in the upper troposphere and their relationship to recent or past convection. The spatial domain of the STRAT (Figure a below) measurements between 80-200 hPa (where there are significant differences between observations and simulated concentrations) is much smaller compared to the other missions. Here we compare with the Pre-AVE mission as an example (Figure b below). The Pre-AVE measurements indicate that there is significant spatial variability in the observed concentrations of CHBr₃, varying from ~0 pptv to >0.4 pptv. The STRAT measurements were obtained in a relatively clean pocket of air that was not recently influenced by convection and therefore show very low CHBr₃ concentrations. Secondly, it is possible that uptake of CHBr₃ on canister surfaces contributed to lower observed bromoform concentrations in the upper troposphere. Though the canisters for STRAT were pre-cleaned with moist air, the treatment of sample canisters for the STRAT mission did not include addition of extra water vapor that became the standard protocol for subsequent missions. The extra water vapor helped minimize adsorptive losses to the canister surfaces.

We agree with the reviewer that since stratospheric observations reported by Sturges et al. (2000) are from the mid- and high latitudes, these mid- and high latitude measurements are not expected to show high CHBr₃ mixing ratios, unlike the tropics. We have deleted the reference to Sturges et al. (2000) in the manuscript.

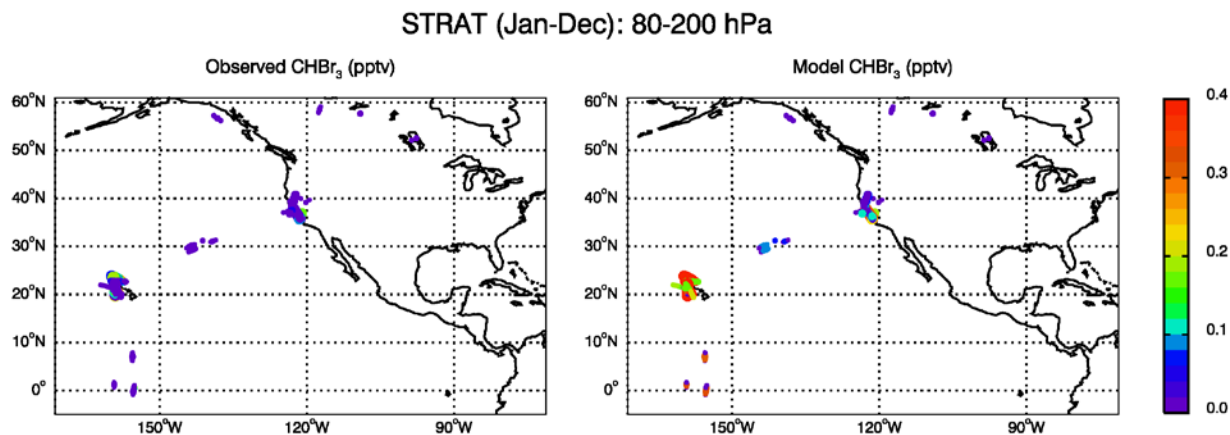


Figure a. Distribution of the observed (left) and simulated (right) CHBr_3 concentrations for the STRAT mission between 80-200 hPa, color-coded according to concentrations and size-coded according to sampling pressure (size increases as pressure increases).

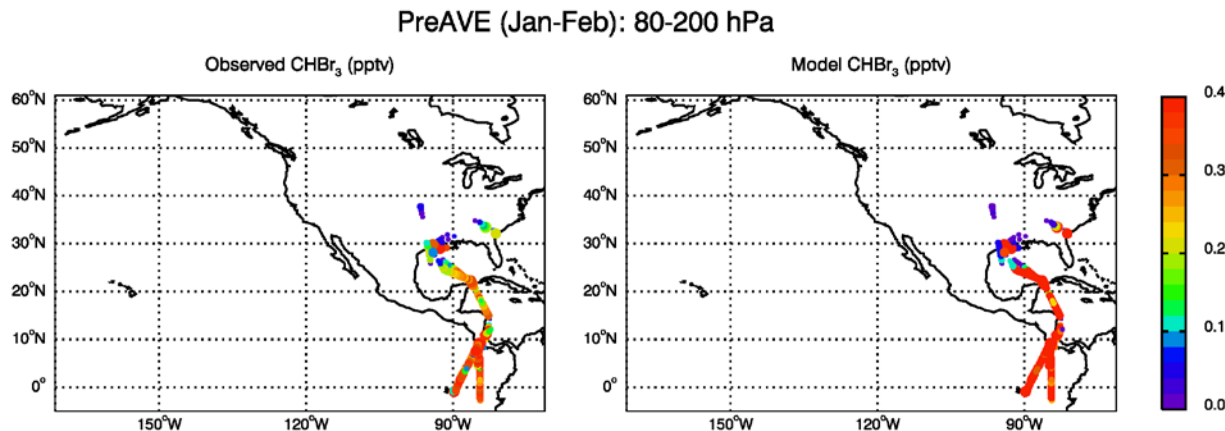


Figure b. Same as Figure a but for the Pre-AVE mission.

p. 23637, Fig. 10: Could you comment on the differences between model and observations of CH_2Br_2 in the lower stratosphere between 100 and 50 hPa seen in Fig. 10?

To address the reviewer's comments, we have examined in detail what might have contributed to the model bias between 100-50 hPa. We have updated Figure 10, showing comparison for individual missions, to better understand the model bias. Figure 10 shows that simulated CH_2Br_2 agrees well with the UT/LS observations below 150 hPa, but is generally too high compared with those between 50-150 hPa, except during Pre-AVE. While the observed CH_2Br_2 show significant interannual variation between missions (varying between 0.3-0.6 pptv at 100 hPa), the simulated CH_2Br_2 vary little from year to year. The model overestimate is likely due to the simplification of using the same zonal-averaged 2-dimensional monthly mean OH fields for each year. However, the fact that the model shows a consistent overestimate between 50-150 hPa during most of the missions suggests our OH is possibly low compared to mean conditions. We have added this in the manuscript.

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

We have added a discussion on possible reasons and implications as suggested.

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

Corrected.

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 Br_y from VLSL delivered into the stratosphere. In particular as you have assumed (a) that all inorganic bromine will be highly soluble and (b) Br_y 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 Hossaini et al.(2009) is slightly out of place here as they don't explicitly consider scavenging. Finally, I suggest that you discuss briefly how your results compare to other published results on the amount of VLSL delivered into the stratosphere and the relative contributions of source gas and product gas injection.

To address the comments from Reviewer #1 and that from Reviewer #2 on convective scavenging of Br_y from VLSL, we have expanded the discussion on the importance of convective transport and dehydration in troposphere-to-stratosphere transport (section 1), how we set up wet deposition in the model (section 2.2), and the implications of our results (section 5). We assume all inorganic bromine is highly soluble and is completely scavenged when condensation occurs in convective updrafts. However, a fraction of the dissolved Br_y is released back to the atmosphere when re-evaporation occurs. When release associated with re-evaporation occurs above the level of neutral buoyancy in the tropical tropopause layer, Br_y can escape to the stratosphere through radiative ascent. Our results suggest that net dehydration during troposphere-to-stratosphere transport occurs slowly so most Br_y that is lofted in convective updrafts is released back to the atmosphere during evaporation, subsequently escaping to the stratosphere. We agree with the reviewer that the reference to Hossaini et al. (2009) is slightly out of place and we have deleted it from the text. We have also followed the reviewer's suggestion and included a brief comparison of our results to other published results on the amount of Br_y^{VLSL} delivered into the stratosphere and the relative contribution of source gas and product gas injection (section 5).