

Evaluating global emission inventories of biogenic bromocarbons.

Response to Anonymous Referee #2

We thank the reviewer for his/her comments. The review comments are repeated below in *italics* and our responses to each are given in **bold**.

The contribution of the very short lived brominated compounds to the stratospheric inorganic bromine load is an interesting and actively debated topic in atmospheric chemistry. The manuscript by Hossaini et al. titled “Evaluating global emission inventories of biogenic bromocarbons” uses currently available emission inventories of CHBr₃ and CH₂Br₂ (two most important brominated VSLs) in a CTM (TOMCAT) and compare simulations with surface measurements from the NOAA global flask network and aircraft data from the recent HIPPO and SHIVA campaigns. This is a well written paper that fits the scientific scope of ACP. The work presented in this paper leads to better constraint on the stratospheric Br_y load from VSLs emissions. The results suggest the number is in the lower end of previous range of estimates at about 4 ppt. My overall opinion about the manuscript is positive. There is one area where the manuscript falls short of my expectations, which I explain in the following paragraph. I would like to see some improvement on that front before it gets published in ACP, although the changes will likely be relatively minor.

Of the four available inventories evaluated in the manuscript, three of them are top-down. While the paper does a very good job pointing out the shortcomings of these top-down inventories in matching the new data, it does not offer enough discussion on why the prior top downs all seem to overestimate CHBr₃ or CH₂Br₃ emissions or both. What went wrong with the previous attempts?

Three of the inventories tested are derived using a top-down approach. Broadly, that is to say they used aircraft observations to infer emissions rather than observed sea-to-air fluxes. There seems to be some consistency in the top-down approach. An example would be for CH₂Br₂ emissions derived in Liang et al. (2010) and Ordonez et al. (2012). Both were formulated using similar aircraft data (i.e. mainly from the same NASA campaigns) and both paint a similar picture of the distribution of emissions (e.g. Figure 3). These inventories are both found to perform well against the HIPPO and NOAA data. The reason the Warwick2011 inventory is larger is likely because it was based on the work of Warwick et al. (2006), in which significantly less aircraft data was used to formulate emissions. It was useful to run simulations with such a larger emission flux in order to assess the plausibility of such emissions. In the revised version of the manuscript we shall discuss more of the discrepancies in the emission inventories that lead to variation in matching the new observations. To note, nothing has likely “gone wrong” with the previous attempts, merely variation in the data used to formulate these inventories leads to variation in the inferred emission fluxes (space and magnitude). In fact, we show that with Liang et al. (2010) in particular (a top-down inventory), we get good agreement in most locations for CHBr₃ and CH₂Br₂. The exception is in the West Pacific, where limited data from that region was used to formulate the emissions.

It is stated in the manuscript that long-term ground based observations, which were lacking in the work that led to the existing top-down estimates, provide better constraints. Surely everyone would agree that more data with better temporal and spatial coverage would provide better budgetary constraints but agreement with aircraft data simulations are not very good either. Some justification is needed on why we should ignore differences on the modelling side, for example. On this note, some brief info on the modelling that went into creating the evaluated inventories should be provided in the manuscript. From what I read, TOMCAT was used in only one of three top-down inventories. Would the current TOMCAT simulations be better at reproducing the data sets used in developing the inventories? Equally relevant, or may be more important, would be a comparison on the data side. Is it possible to make direct comparisons between the measurements used in previous works and the data presented here? Could there be issues regarding temporal variability or may be systematic biases between data sets?

Aircraft observations alone only provide a “snap shot” during a relatively small period of (campaign) time. The long-term observations from NOAA provide valuable information. What is required is a

multi-model inter-comparison in order to assess if the emission fluxes found to perform well in TOMCAT also perform well in other models. This, along with assessing the temporal variability in observational data, will be assessed as part of the TRANSCOM-VSLS model inter-comparison project (introduced in conclusions) but is beyond the immediate scope of this paper. In the revised manuscript, we shall include more discussion on how the present inventories were formulated and to what degree the performance of each may be model-dependant.

Some minor comments:

Pg.12489, lns. 14-16: I don't fully understand what is meant by "surface" as it relates to lifetime and the phrase "a global/seasonal mean photolysis rate"? There is another slash on line 16. I think they can use "and" "instead."

For VSLS, it is common to quote a "local lifetime" that assumes a [OH] of 1E6 molecules cm⁻³ and a temperature typical of the surface (i.e. 275K), see WMO (2010).

Pg.12491, ln. 3: May be a reference needed after "TransComCH4".

The missing reference will be included.

Pg. 12496, lns. 5-13: Seasonal cycle in the SH is also driven by photochemistry?

Yes, a strong seasonal cycle at SPO is apparent and is likely due to photochemistry.

Pg. 12507, ln. 9: The end of the sentence, starting with "..., which..." is unnecessary (stated before). There is an "also" at the very end, probably leftover text from editing.

Ok, we shall amend accordingly.