

Dear Referee #2

We thank the referee for his/her valuable comments, which helped to improve the manuscript. We have added a more detailed descriptions of the model and comparisons with other observational data as well as other model studies. The changed parts in the manuscript are marked in red. The following is a point-by-point response.

**Anonymous Referee #2**

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**The authors present an interesting model study on the spatial variation of atmospheric CHBr<sub>3</sub> concentrations. Overall the paper is clearly written, and easy to read. While I agree their main conclusion that the spatial distribution is a result of the interplay of transport and emissions (and chemical depletion), I think a major revision is needed before it can be accepted for publication. My major concerns are:**

**1. There is no sufficient description on their transport model. The FLEXPART model has been widely used in various researches. But for CHBr<sub>3</sub> simulation, the reader still wants to know the important details, including its description on the air-sea exchange, and on the oxidation scheme etc.**

Answer: More details of the chemical decay (loss process), air-sea flux, and calculations of mixing ratios have been added in section 2. We revised the manuscript by adding:

“The air-sea flux was obtained from the transfer coefficient ( $k_w$ ) and the concentration gradient ( $\Delta c$ ) between water concentration and the theoretical equilibrium water concentration (see details in Fiehn et al., 2017 and reference therein):

$$F = k_w \cdot \Delta c \quad (1)''$$

in lines 118-124.

“... Trajectories released from the global ocean surface or along the cruise track carry the amount of CHBr<sub>3</sub> prescribed by the respective emission scenario. Chemical decay of CHBr<sub>3</sub> was accounted for by:

$$m(t + \Delta t) = m(t) \exp(-\Delta t/\beta) \quad (2)$$

where  $m$  is the particle mass,  $\beta=T_{1/2}/\ln(2)$  is the  $e$ -folding lifetime of CHBr<sub>3</sub>, and  $T_{1/2}$  is the half-life of CHBr<sub>3</sub> (Stohl et al., 2005). In our study, a half-life of 17 days ( $e$ -folding lifetime of 24 days) is prescribed to CHBr<sub>3</sub> during all runs (Montzka and Reimann, 2011).” in lines 150-156.

“Output data in form of CHBr<sub>3</sub> volume mixing ratios (VMR) available at a user-defined grid, were calculated by:

$$VMR = \left(\frac{C_T}{\rho_a}\right) \cdot \left(\frac{m_a}{m_T}\right) \quad (3)$$

where  $C_T$  is the CHBr<sub>3</sub> mass concentration,  $\rho_a$  is the density of the air, and  $m_a$  and  $m_T$  are the molecular weight of air and CHBr<sub>3</sub>, respectively.

For each grid cell, the CHBr<sub>3</sub> mass concentration is given by:

$$C_T = \frac{1}{V} \sum_{i=1}^N m_i f_i \quad (4)$$

with  $m_i$  being the mass of CHBr<sub>3</sub> for particle  $i$ ,  $f_i$  the mass fraction of CHBr<sub>3</sub> of particle  $i$  attributed to the respective grid cell,  $N$  the total number of the particles, and  $V$  the volume of the grid cell (Stohl et al., 2005). We run FLEXPART in the non-domain filling mode, therefore the particle distribution is not correlated with air density. Particles, and thus bromoform, can accumulate in regions of low wind speeds where the relatively long residence time allows that oceanic emissions constantly add new particles. Similarly, particles can accumulate in regions of convergence where horizontal inflow pools marine boundary layer air from different regions..” in lines 185-198.

**2. There is no comparison of their simulations with other models, or more importantly with observations. There are questions on how realistic their simulations are. For example, the model shows large areas with very low CHBr<sub>3</sub> concentrations (<0.1 pptv), particularly at the 5km level (Figs.8 to 10). They appear inconsistent with aircraft measurements such as the recent CAST and CONTRAST campaigns.**

Answer: The simulations in our paper based on the uniform background emissions, only include open ocean CHBr<sub>3</sub> emissions and do not take coastal emissions into consideration. As coastal and shelf emissions contribute significantly to atmospheric CHBr<sub>3</sub> mixing ratios, we do not expect our runs to give realistic mixing ratios. The main point of our simulation is not to determine the overall amount of atmospheric CHBr<sub>3</sub>, but to test whether the signals of localized elevated CHBr<sub>3</sub> emissions (hot spots) could be distinguished from those of background emission. To clarify the main point and avoid confusion, we rearranged and rewrote some sections of the manuscript. We added a paragraph in the discussion section to compare our simulations with some aircraft campaigns and model studies explaining differences between the two based on our approach. Several new citations (Harris et al., 2016; Pan et al., 2016; Butler et al., 2018) are also included.

“Our approach requires that we isolate uniform background CHBr<sub>3</sub> emission from coastal and shelf emissions, which can be significant (Fuhlbrügge et al., 2016; Fiehn et al., 2017) and would lead to higher atmospheric abundances. In consequence, we expect the background CHBr<sub>3</sub> mixing ratios inferred from our simulations to be smaller compared to observations and other modeling studies. In the Western Pacific (TransBrom), our simulated background mixing ratios at 5 km range from 0.0-0.4 ppt (Fig. 9-11). Measurements from aircraft campaigns in this region, CAST (Harris et al., 2016) and CONTRAST (Pan et al., 2016), show higher CHBr<sub>3</sub> mixing ratio of 0.03-0.79 ppt and 0.20-1.127 ppt between 4-6 km. Other model studies (e.g. Hossaini et al., 2016; Butler et al., 2018) based on CHBr<sub>3</sub> emission scenarios that include coastal and open ocean sources (e.g. Liang et al., 2010; Ordóñez et al., 2012; Ziska et al., 2013) also suggest the average CHBr<sub>3</sub> mixing ratio over 0.5 ppt in this region. ” in lines 423-432.

**3. More detailed analysis is needed. At some places, the paper tends to establish the correlation between the CHBr<sub>3</sub> distribution and the (snapshot of) wind fields. Considering the lifetime of CHBr<sub>3</sub>, such correlations are often not obvious, as**

**demonstration by their own results. Tagged simulations (for example, Butler et al., 2018) may help the reader understand the complexity, particularly for the emission ‘hotspots’.**

Answer: To show the correlations in a clearer way, we added a statistical analysis of the CHBr<sub>3</sub> distribution by regions of convergence/divergence and wind speeds. The results suggest that higher mixing ratios tend to appear in the regions of convergence or lower wind speed. The statistical results are shown in new Figure 5.

The manuscript is revised by adding “In order to validate the hypothesis, we show a violin plot of regional background CHBr<sub>3</sub> mixing ratios related to convergence/divergence, and to the wind speeds averaged over each simulation period in Fig. 5. For the TransBrom case, the averaged ranges of mixing ratios in regions of convergence and divergence (Fig. 5a) go up to 0.7 ppt and 0.5 ppt, respectively, with interquartile ranges of 0.1-0.35 ppt and 0.05-0.21 ppt. Probability of mixing ratios larger than 0.2 ppt is much higher for regions of convergence compared to regions of divergence. Meanwhile, in the regions grouped by wind speed (Fig. 5b), higher CHBr<sub>3</sub> mixing ratios are more likely to occur in regions with lower wind speeds (i.e. in the regions of 0.0-5.0 m/s, mixing ratio go up to 0.65 ppt, while in the regions of 10-15 m/s, mixing ratio go up to 0.25 ppt). Similar distributions also occur for the SHIVA case. During the OASIS case, the CHBr<sub>3</sub> mixing ratios are much smaller than for the other two cases due to stronger winds. Highest mixing ratios (~0.15 to ~0.2 ppt) are found in the regions of convergence (Fig. 5e). However, higher mixing ratios are also generally found in the regions of higher wind speeds (Fig. 5f), as the regions of convergence locate in the regions of high wind speed during the OASIS case. The distributions suggest that in general higher CHBr<sub>3</sub> mixing ratios tend to occur in the regions of convergence or lower wind speed, with the exception of the OASIS case where extremely high winds occurred and coincided with regions of convergence.” on lines 239-253.

Thank you again for all your comments, hope the revised manuscript has addressed your concerns.