

Response to Reviewer 2

We thank Reviewer #2 for his/her comments leading to a significant improvement of the manuscript.

This response is organised with the reviewer's comments in black bold font, the authors' answers in black normal font and the text added in the revised manuscript in red font.

Note that following reviewer #1's comment:

- figure 2 has been removed but the flight trajectory is now included in figure 1
- a new figure 7 have been included showing a direct comparison between the simulation and the measurements on a map.
- the naming of the modelled systems has been changed to include their latitude (Mod_Conv1, Mod_Conv2 and Mod_Conv3 replaced by Mod_Conv_4.35N, Mod_Conv_3.75N, and Mod_Conv_5.4N, respectively)
- figure 5 showing the model cloud top has been replaced by an estimate of the brightness temperature from the cloud top pressure (new figure 4) to provide a direct comparison to the observed brightness temperatures (figure 1)
- because section 7 was very short, we have now merged section 7 with section 6 and made it clear the final paragraph presents a short outlook

To improve the clarity of the manuscript, the discussion section (4.3.4) is now split into two sections with the comparison to Marécal et al. (2012) being now in a separate section (new section 4.3.5).

The paper describes a cloud-scale modeling study to investigate convective transport of bromoform (CHBr₃) and its product gases. The model is applied to a case study along the west coast of Borneo that was sampled with aircraft measurements, which provide a means to evaluate the model. The major findings are that there is good agreement of CHBr₃ mixing ratios between model and observations in the boundary layer and reasonable agreement in convective outflow regions. Analysis of the bromine speciation in convective outflow shows ~85% of the Br is from CHBr₃, < 10% from inorganic product gases (HBr, BrO, HOBr, Br, Br₂, BrONO₂), ~2% from organic product gases (brominated peroxides and carbonyls). The paper finds that the inorganic product gases are dominated by HBr, which is highly soluble and quickly removed by the convection. The paper suggests that the high HBr is a result of the low O₃ environment (O₃ < 20 ppbv) that favors production of HBr from Br + HCHO and Br + HO₂ over production of BrO from Br + O₃. Without BrO production, HOBr also has low mixing ratios. Further, aqueous phase reaction between HOBr and HBr is fast, further limiting HOBr within convection.

This study provides fundamental knowledge on the processing of Br compounds in tropical convection, which is important to apply to global models that examine pathways of Br compounds to the stratosphere where Br plays a critical role in ozone chemistry. The paper covers several topics without fully justifying why each topic is addressed. My main concern is that the main points are not concisely given (e.g. in the abstract or results section) but are written along with other points that cause a loss of clarity in the story. For example, why do we need to know the convective transport efficiency? Secondly, why is it important to know if CHBr₃, HBr, BrO, or organic Br is the bromine compound transported to the upper troposphere?

The paper has been revised to make the analysis of the results more organized and clearer (see answers to specific comments below). We have made clear that the comparison of the transport efficiency is a mean to evaluate the simulation compared to observations. We have also largely improved the discussion on the bromine partitioning in particular making clearer its aims.

In addition, the discussion focuses on the regional chemistry that the convection forms in. The discussion makes sense but is not supported with figures showing rates of reactions.

As suggested, we have now given details on the reaction rates to support our analysis (see answers to specific comments).

Specific Science Comments

1. The abstract needs to be improved by making the existing text be more concise, stating more clearly that low O₃ levels lead to HBr gas-phase production, and adding comments on the role of aqueous chemistry.

We have now shortened the abstract to make it more concise by removing unnecessary details repeated later in the paper. We have also highlighted the link between low ozone and HBr in a clearer way.

2. Line 102 in Introduction. I do not think it was explained what the limitations are of the previous studies. There should be a few sentences clearly stating these limitations in the Introduction and again in the Discussion.

The previous studies were only idealized cases that used induced convection in the simulations by introducing artificial atmospheric perturbations and used various simplifying assumptions. We have now highlighted this as their main weakness and limitation in the introduction:

“The previous studies of VLS chemistry and transport at the convective scale were only idealised cases that used a set of simplifying assumptions (e.g., no emissions, constant vertical profiles for initial conditions, and no synoptic scale meteorological forcing) and artificial perturbations to the modelled atmosphere to induce their simulated convection. Thus, these cases were not realistic and it would not have been relevant to compare them to observations. We wish to expand upon that previous work, e.g., Marécal et al. (2012), by carrying out a real-world case study.”

We then repeat some of this information in the discussion in more detail within the new section 4.3.5. This also covers responses to comments made by reviewer 1.

3. Line 153 states that the CHBr₃ measurements were performed by the GHOST instrument. Please add information on what the technique for these measurements is, as well as its detection limits and uncertainties.

We have now added text in section 4.2 explaining that *“The GHOST instrument is a gas-chromatograph mass spectrometer and had an error of $\pm 17.7\%$ that was primarily driven by uncertainties in the gas standard.”*

4. Since the GHOST instrument is a GC/MS technique, are there other trace gases that it measured that are useful for this analysis, such as other Br-containing trace gases?

The GHOST instrument does measure other bromocarbons, but these compounds are not included in our chemical mechanism and so these additional observations cannot help our analysis. Beyond the GHOST instrument, although it was initially planned in SHIVA to measure other bromine species, logistical problems with aircraft availability created a tight limitation on the payload for the mission. As a result, it was not possible to take chemical ionization mass spectrometry instrumentation onboard, which would have been able to measure some of the inorganic components.

5. Line 223. I noticed that the chemistry listed in the supplement does not include Br-Cl reactions, e.g. $\text{Cl} + \text{CHBr}_3 \rightarrow \text{HCl} + \text{CBr}_3$. What impact would Cl chemistry have on the results provided in this paper?

A sensitivity analysis on the impact of Cl chemistry was done in Marécal et al. (2012). They showed that chlorine chemistry has only a negligible impact on the production of Br_x . This is why we have not included Br-Cl reactions in the chemistry scheme used in the paper. This information is added in the revised manuscript.

“Note that bromine-chlorine reactions have not been included in the chemistry scheme since Marécal et al. (2012) showed that it has a small impact on the production of Br_x .”

6. Does the model include direct uptake of Br compounds onto ice? It may not be important for the species considered (except BrONO_2 ; Fernandez et al., 2014, ACP), but I suggest mentioning this process.

There is no direct uptake of Br compounds onto ice. Br is transferred to ice through liquid-to-ice processes (like riming) using retention coefficient as explained in section 3.1. These processes are dominant in the formation of ice hydrometeors in deep convective systems.

“This approach represents liquid-to-ice processes like riming that are the dominant process for the formation of ice hydrometeors in convective clouds.”

Furthermore, we find that HBr is almost entirely removed at an altitude in the convection column where hydrometeors exist only in solid form. Therefore, we do not expect an important effect of direct uptake onto ice as also discussed in Marécal et al. (2012) Sect. 3.2.1.

“The uptake of bromine species onto ice hydrometeors is not represented as it was found in Marécal et al. (2012) to not have an important effect on bromine removal.”

7. Lines 245-249. Could you provide a little more information on the emissions? Do the emissions vary temporally? If not, what time-span is the average for?

We have used a diurnal variability on the emissions linked to solar zenith angle such that emissions peak at solar noon. The mean emission in $\text{pmol m}^{-2} \text{h}^{-1}$ is equal to that shown in Fig. 3. We have added this information in the text.

“A diurnal variability linked to solar zenith angle is applied to these emissions such that they peak at solar noon.”

8. Line 298. What is the vertical resolution of the model in the upper troposphere? Does the vertical resolution affect the cloud top height estimate? Since the results show a fairly good comparison, I guess not, but I suggest thinking about the uncertainties in the model cloud top height if $\text{dz} > 500 \text{ m}$.

At the altitudes near the cloud top height the model vertical resolution is $\sim 300 \text{ m}$. This does mean that the precision of the model's cloud top height estimates were equivalent to this resolution. Note that, on the advice of reviewer #1 we now use a model estimated cloud brightness temperature as a measure of cloud top height. This was preferred because we show satellite observations of cloud top brightness temperature. This does not change the results that still show a good agreement between the model and the observations.

9. Line 328. Are there any temporal uncertainties to consider? I imagine the statistical approach removes effects of poor timing of the convection, but does it also reduce uncertainties in relation to timing of emissions (if there is a diurnal profile) or photochemistry?

We think the main effect is in removing effects of different timing of the convection. It is harder to say anything concrete about the emissions uncertainties since little is known about the short term variability of the emissions. Temporal emission errors would mainly effect the accumulation of bromoform in the boundary layer prior to convective entrainment and the amount of bromoform lofted in the convective column. The statistical approach has more an effect of temporally smoothing the detrainment and so this choice does not remove emission error.

10. Line 330. I first saw the equation given on line 333 in Cohan et al. (1999) JGR (please cite them). This equation assumes that there is no mixing of air at different altitudes between cloud base and cloud top, i.e. it is a two-component mixture model. During the past decades, others have used modified versions of this type of equation. For example, Borbon et al. (2012) JGR applied a three-component mixture model, while Yang et al. (2015) JGR applied a four-component mixture model, and Fried et al. (2016) JGR applied a 10-layer mixture to account for entrainment of air between cloud base and cloud top. The multi-layer approach is useful for conditions when the vertical profiles vary with altitude in the clear air. It would be good to see the clear-sky vertical profile for CHBr₃ to show its variability with altitude. Is CHBr₃ sufficiently close to zero or non-varying with altitude such that entrainment of air between cloud base and cloud top can be ignored?

Reference to Cohan et al. 1999 added. The reason we have used the approach based on a two-component mixture model is that we wanted to evaluate the simulation by making a comparison with Krysztofiak et al. (2018) study that chose this approach. We agree that a multi-layer approach is better for vertical profiles varying with altitude. Looking at the background CHBr₃, we find that there is very little variability from 2 km to the upper troposphere (CHBr₃ 0-0.5 pptv). Between 400m and 2km altitude, CHBr₃ concentrations are slightly affected by the lowest levels but are still below 1 pptv.

Also following reviewer #1's comments we have revised the introduction of section 4.2 and added more explanation of f (air fraction transported from the BL to the UT).

“Before discussing the results of the simulated chemistry in detail (section 4.3), we evaluate if the simulation gives reasonable results for CHBr₃ concentrations and for convective transport efficiency compared to the aircraft observations.

We firstly use statistical characteristics for this comparison. We choose this approach for two reasons. First because of differences in location and timing between the observed and simulated convection events, and, second, because of spatial uncertainties in the emission inventory used in the simulation. This approach allows a clearer comparison of the observations and simulation by removing effects arising from inherent time and spatial uncertainties.

In order to compare the convective transport efficiency between the observed and simulated systems we follow the approach proposed by Cohan et al. (1999) and used by Bertram et al. (2007). To estimate the air fraction, f , originating from the boundary layer (BL) and transported by convection we use the relationship from Cohan et al. (1999):

$$[X]_{UTconv} = f \cdot [X]_{BL} + (1 - f) \cdot [X]_{UTnoconv}$$

where the mean mixing ratios in the boundary layer, the upper troposphere within the convective systems, and the upper troposphere in the vicinity but outside the convective systems are represented by $[X]_{BL}$, $[X]_{UTconv}$, and $[X]_{UTnoconv}$, respectively. f ranges from 0 to 1 with large values corresponding

to an efficient convective transport of air masses from the boundary layer to the upper troposphere. This formulation of f is chosen because it was recently applied to the SHIVA aircraft data (Krysztofiak et al., 2018). It relies on the assumption of a low variability of background concentrations with altitude, which is fulfilled for CHBr_3 in our case study (not shown). Previous studies based on observations and reported in Krysztofiak et al (2018) provides estimates of f in the range 0.17 to 0.36.”

11. Lines 388-395. Could you specify where the vertical cross section is located. Is it an average across the anvil, or a line down the center of the anvil?

The cross section is taken as close to the centreline of the convective cell as possible. We have now explained this in the text.

“The cross sections in each case run as close to the centreline of each convective system as possible.”

12. Section 4.3.2. Figures 9 and 10 show values based on Br pptv and percent contribution to total Br. Somehow the text gets a bit confusing. Is it possible to organize the text a little differently, such as by region, which would better align with the Conclusions section. For example, In the UT convective outflow CHBr_3 contributes 85%, inorganic PGS < 10%, and organic PGs ~2% to total Br. In the boundary layer, CHBr_3 contributes In the 1-4 km layer, CHBr_3 contributes

This section has been organised following this suggestion making it clearer

“Figs. 9 (d), (e), and (f) show that there are relatively low levels of inorganic bromine (Br , Br_2 , BrO , HOBr , HBr , BrONO_2) concentrations in the boundary layer even in the areas not directly affected by convective precipitation with values typically in the range of 0 to 0.4 pptv Br, i.e., <5 % contribution to the total Br mixing ratio (Figs. 10 (d), (e), and (f)). The highest simulated inorganic bromine mixing ratios (0.3-0.4 pptv Br) in the boundary layer occur to the west of the Mod_Conv_5.4N system still only contributes <10 % to the total boundary layer pptv Br (see Fig. 10 (f)). This spatial variability in the boundary layer inorganic bromine mixing ratios around each convective system arises due to differences in precipitation location and timing over the course of the simulation prior to November 19th, 2011. Precipitation events occurring in the two preceding days deplete the boundary layer of inorganic bromine due to washout (analysis not shown). In the boundary layer, organic PGs (CHBrO , CBr_2O , CHBr_2OOH , and CBr_3COOH) concentrations are up to 0.2 pptv Br (up to 10% contribution to total bromine but very locally) and are formed due to CHBr_3 photochemical loss (Figs. 9 (g), (h) and (i) and 10 (g), (h) and (i)).

Air masses in the convective column itself and convective outflow are almost entirely depleted of inorganic bromine with mixing ratios of <0.1 pptv Br and with contributions to the total Br mixing ratio well below 5%. There, organic compounds are being driven from the low levels up to the upper troposphere in the main ascent and the outflow and show enhanced mixing ratios within the convective column compared to the free troposphere (Figs. 9 (g), (h), (i)). Still organic PGs have a contribution to the total bromine only up to ~4%.

In the free troposphere, inorganic and organic bromine concentrations are enhanced between 1 and 4 km to the west of each convective system (Figs. 9 (d), (e), (f), (g), (h), (i) and 10 (d), (e), (f), (g), (h), (i)). There, total inorganic (resp. organic) PGs peak up to 1 pptv Br (resp. 0.2 pptv Br), which constitutes a portion up to 45% (resp. 5%) of the total Br mixing ratio. Among the three convective systems, Mod_Conv_5.4N exhibits the highest concentrations of the organic PGs.

Above 4 km altitude in convection-free areas, inorganic bromine is mainly in the 0.2-0.4 pptv Br range (15-35% contribution to total Br) that is higher than within convection (Figs. 9 (d), (e), (f) and 10 (d), (e), (f)). There, organic PGs have low concentrations (0.02-0.03 pptv Br) and contributes only to 1-3% (Figs. 9 (g), (h), (i) and 10 (g), (h), (i)).”

13. Line 488. It is suggested that reactions 2 and 3 could be important routes for HBr formation. Reaction 2 is Br + HCHO. Does HCHO come from CH4 or are there VOCs, such as isoprene, contributing to its mixing ratio? What is the typical reactivity for reactions 1 to 3 for the study area? It would help to see reaction rate constants and typical O3, HCHO, and HO2 mixing ratios.

There are other VOCs present beyond methane, and biogenic isoprene emissions contribute towards the HCHO mixing ratio. We agree that it would be useful to list the reactivity of reactions 1-3. We have added table 4 now to show the typical HO₂, O₃, and HCHO mixing ratios along with the rate constants for these reactions and the resulting reaction rates. We include a new paragraph discussing this table and the implications of these results. Having examined the rates of the various equations in more detail we have now added a discussion of a reaction that was not previously mentioned (BrO + NO), which plays a role in helping to suppress BrO levels. We considered plotting the cross-section structure of the reaction rates for each compound, but this rapidly becomes very complicated to analyse and requires extensive explanation. Table 4 and the accompanying explanation offer a compromise that conveys the essential information in a condensed form. Additionally, we used mixing ratios in the lowest part of the troposphere (200 m) to construct Table 4 because it is the mixing ratios in these air masses that become entrained within the convective systems.

14. Line 505. Again, it would help to know the reaction rate constant and reactivity of reaction 7. Marecal et al. (2012) report 1.6x10¹⁰ M⁻¹ s⁻¹, which is indeed high but is the rate high as well when compared to competing gas-phase reactions? It would be good to see figures of reaction rates supporting the discussion on what reactions dominate.

Following this query we have compiled all of the relevant reaction rate constants and reaction rates discussed within section 4.3.4 into a new table (Table 4). The data in Table 4 are selected for a point in the boundary layer at 200 m altitude in an area to the north of Mod_conv5.4N. This location is part of an air mass that becomes entrained in Mod_Conv5.4N and is representative of the typical air masses entrained in the three systems. Note that we have added three more reactions into the discussion to give a more complete explanation (BrO + NO, and the photolysis of both HOBr and BrONO₂), and as a result R7 is now numbered as R10. As the reviewer points out, the reaction rate constant we have used for R10 (HOBr+HBr) is calculated in the same way as in Marecal et al. (2012). Indeed, R10 has a very fast rate constant, which has implications for HOBr lifetime within cloud and rain droplets (see response to next query). However, despite the fast rate constant, the overall rate of R10 in the example given in Table 4 is low and we have determined this is because R10 is strongly limited by the availability of HOBr. R10 only becomes important in a few areas where background levels HOBr is elevated as a result of higher background levels of BrO. The new discussion highlights importance of R2 (Br + HCHO) for leading to HBr formation, R4 (BrO + HO₂) for removing BrO, and R9 (HOBr + hv) for recycling HOBr back to Br and for keeping HOBr mixing ratios very low.

Since we substantially rewrote section 4.3.4 amounting to nearly 3.5 pages of text we have not copied the full text here and quote the most relevant passages along with Table 4.

“Within the photochemical scheme of C-CATT-BRAMS, bromine atoms can react in the gas phase in one of three ways **via R1, R2, or R3 (shown in order of decreasing reaction rate).**

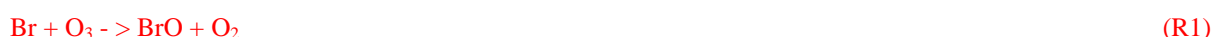


Table 4: Rate constants and reaction rates at a point in the marine boundary layer (200 m, 988.7 mb and 298.7 K)

within an air mass being advected into the convective updraft of Mod_Conv_5.4N (5.8 N 115.5 E). This was done based on chemical species' mixing ratios at this location for all of the reactions (R1-R9) discussed in Sect. 4.3.4. This was in a region with no cloud or rain. For R8 and R10, the reactions taking place in the aqueous phase, we select some representative rain and cloud water mass mixing ratios (0.5 g kg⁻¹ and 0.1 g kg⁻¹, respectively) to demonstrate the reaction rates in the presence of liquid water condensate. In the case of R10 the hypothetical reaction rate is calculated using the aqueous concentration of HOBr at Henry's Law equilibrium, and due to the extremely high solubility of HBr it is assumed that all of its gas phase mixing ratio is dissolved in solution. For both R8 and R10 the reaction rates in cloud and rain droplets are shown separately, but in the case of R10 they cannot be combined additively.

Reaction	Mixing ratio of 1 st reactant	Mixing ratio of 2 nd reactant	Rate constant	Reaction rate / molecules cm ⁻³ s ⁻¹
R1: Br + O ₃	0.08 ppqv	13.1 ppbv	1.17×10^{-12} molecules ⁻¹ cm ³ s ⁻¹	678
R2: Br + HCHO	0.08 ppqv	3.7 ppbv	1.17×10^{-12} molecules ⁻¹ cm ³ s ⁻¹	191
R3: Br + HO ₂	0.08 ppqv	37.2 pptv	1.70×10^{-12} molecules ⁻¹ cm ³ s ⁻¹	2.8
R4: BrO + HO ₂	0.5 ppqv	32.5 pptv	1.59×10^{-11} molecules ⁻¹ cm ³ s ⁻¹	185
R5: BrO + NO ₂	0.5 ppqv	4.6 pptv	2.79×10^{-12} molecules ⁻¹ cm ³ s ⁻¹	4.0
R6: BrO + NO	0.5 ppqv	0.6 pptv	2.10×10^{-11} molecules ⁻¹ cm ³ s ⁻¹	3.8
R7: BrONO ₂	0.8 ppqv	-	9.45×10^{-4} s ⁻¹	18.4
R8: BrONO ₂	0.8 ppqv	-	3.73×10^{-3} s ⁻¹ (rain: 1.33×10^{-4} , cloud: 3.60×10^{-3})	72.6
R9: HOBr + hv	3.6 ppqv	-	1.63×10^{-3} s ⁻¹	139
R10: HOBr + HBr	3.6 ppqv	0.23 pptv	5.44×10^{-6} molecules ⁻¹ cm ³ s ⁻¹ (rain) 2.72×10^{-5} molecules ⁻¹ cm ³ s ⁻¹ (cloud)	0.22 (rain: 0.18, cloud: 0.04)

Bromine radicals can react with ozone via R1 to form BrO, which is normally the dominant reaction pathway for Br in the troposphere and this is the case here too. In addition, bromine radicals can react with HCHO or HO₂ to form HBr, but combined the rate of HBr formation via R2 and R3 is less than a third of the production rate of BrO. However, under relatively low ozone conditions (13.1 ppbv in this example) and in the presence of relatively high HO₂ (37.2 pptv) and only low NO_x levels (e.g., 4.6 pptv of NO₂ and 0.6 pptv of NO), as we find in our simulations, the reactions between BrO and (in order of decreasing importance) HO₂ (R4), NO₂ (R5), and NO (R6) are enough to suppress BrO mixing ratios to only negligible levels.



Combined R4, R5, and R6 suppress boundary layer BrO mixing ratios down to very low levels (0.5 ppqv) within this example air mass during the daytime. During the night BrO production will shut off, and residual NO as well as NO₂ react with BrO resulting in the latter's complete removal.

R4 leads directly to modest HOBr formation. R5 leads to BrONO₂ formation but its mixing ratios are also kept low by photolysis in combination with hydrolysis within cloud and rain droplets.



The hydrolysis of aqueous phase BrONO₂ R8 is another pathway leading to HOBr formation, which is dependent on the presence of condensed moisture (note that for numerical reasons, in the model, the HOBr and HNO₃ from R8 are produced in the model in the gas phase as their partitioning into the aqueous phase is determined by Henry's Law and air-liquid diffusion limited uptake). However, despite modest formation rates of HOBr via R4 and R8 the mixing ratios of HOBr are suppressed to relatively low levels during daytime by photolysis.



Meanwhile, HBr (formed by R2 and R3, albeit relatively slowly) only has a very slow rate of photolysis and it is thus the single inorganic bromine compound with a lifetime long enough to allow accumulation in the gas phase under these conditions. HOBr and BrONO₂ are the next most abundant inorganic PGs, but their lifetimes are kept low during the daytime by photolysis. If background ozone levels were higher it would allow greater formation rates of BrO and in turn HOBr and BrONO₂ since BrO is the start point in the reaction pathways for both species."

15. Line 521. It mentions the very short residence time due to falling hydrometeors. Can you calculate the residence time? I would suggest looking at Bela et al. (2018) JGR who discuss time an air parcel spends in contact with liquid water in convection (ranging from weak convection to severe convection). They also showed via vertical profiles that highly soluble trace gases (like H₂O₂) are depleted rapidly below the freezing level, whereas less soluble gases will be lofted higher.

Following this query (and the one above) from the reviewer we spent some time trying to understand the residence time versus the reaction rate for the HOBr + HBr reaction (now R10). As a result, we realized that the original explanation in the paper, i.e., that the residence time was shorter than the lifetime of HOBr or HBr in aqueous solution, was incorrect. While unfortunately we cannot calculate the residence time, we were able to determine that the lifetime of HOBr in the presence of 0.1 g kg⁻¹ cloud and 0.5 g kg⁻¹ (corresponding to the active convective center in the BL) and 0.2 pptv HBr (dissolved) was very short at 2×10^{-2} s such we can be confident it is much shorter than the residence times of rain or cloud droplets. We have added the following text though to draw a contrast with Bela et al. (2018):

“The residence time of cloud and rain droplets in the atmosphere can impact aqueous phase chemistry in cloud and rain droplets. For other aqueous chemistry systems occurring in cloud and rain droplets the rate of chemical reactions could occur slower than it takes for cloud or rain droplets to fall leading to wet scavenging of the chemical species involved. Bela et al. (2018) report an example of aqueous phase chemistry in convective cloud and rain where the wet scavenging removal of H₂O₂ was found to be much faster than its production via aqueous phase chemistry in cloud and rain. By contrast, the rate constant for R10 is approximately 10⁷ times faster than the rate constants involved in the formation of H₂O₂ described by Bela et al. (2018), the lifetime of HOBr in solution is approximately 2

$\times 10^{-2}$ s as a result. Thus, the residence time of cloud and rain droplets is not a limiting factor for R10.”

16. Lines 524-557 discuss how the current results compare to those by Marecal et al. (2012). Could the authors make clear what are the differences between the model configuration by Marecal et al. and this paper? I found two comments on the differences, 1) running real meteorology (and thus global model generated initial conditions) and running ideal meteorology, and 2) running a case for Darwin versus Borneo where emissions, photolysis rates, OH mixing ratios can be different. Are there other differences? What about the information in the “new chemistry” section?

We have now added the following text to explain the differences with the model configuration in Marecal et al. (2012).

“The C-CATT-BRAMS model configuration in Marecal et al. (2012) differs from the setup used in this study in the following ways:

- The model domain was set to be at the same latitude and longitude as Darwin (Australia).
- There were no emissions of CHBr_3 or any other VSLs. The only source of CHBr_3 was from a 2 km homogenous layer above the surface set as an initial mixing ratio of either 1.6 pptv or 40 pptv in the two scenarios.
- Marecal et al. (2012) used a chemical mechanism that did not include a representation of non-methane hydrocarbon chemistry. This chemistry was added in this study by including the ReLACS chemical mechanism.
- Apart from CHBr_3 set in the lowermost 2 km, the model initial and boundary conditions for the chemical species were defined from a single vertical profile from the MOCAGE CTM from over Darwin. The mixing ratios of all PGs were initialized at 0 pptv in all model grids and layers.
- The meteorology in Marécal et al. (2012) involved an initial setup that applied a single vertical profile of meteorological conditions throughout the entire horizontal domain. The vertical profile was defined from a radiosonde profile obtained above Darwin corresponding to November just prior to the main wet season. Convection was artificially forced in the simulation by introducing a perturbation in the lower model layers of increased temperature and humidity.”

17. Line 542. For the discussion of differences due to Darwin and Borneo settings, the lifetime of CHBr_3 is discussed. These lifetimes are > 15 days. How do the changes in chemical lifetime affect the results of convective transport, which occurs in < 1 hour and is the topic of the paper?

The C-CATT-BRAMS simulation is run for almost 48 hours prior to the onset of convective activity while the TOMCAT simulation used for background and initial conditions was run for 3 years prior to the 19th November 2011. The lifetime of CHBr_3 during this preceding period affects the abundance of it and its PGs as well defining the relative composition of the different PG components. So while the typical transport time within the convective systems is less than one hour, the lifetimes with respect to photolysis and reaction with OH, both do play an important role in defining the tropospheric composition of CHBr_3 and its PGs prior to convection. We now make this point a bit clearer by adding the following text:

“The CHBr_3 lifetimes with respect to photolysis and OH are important for defining the relative partitioning of bromine between CHBr_3 and its different PGs prior to the convective activity on the 19th November.”

18. Line 557. Isn't O₃ low in both Darwin and Borneo? I do not see why low O₃ would cause differences between the results from Marecal et al. and this paper.

The intention of this sentence was to draw a parallel with the ozone levels along the coast of Borneo and in the simulation near Darwin in Marécal et al., 2012. However, due to the placement of the sentence and the overall context of the paragraph, we realise this is not very clear. We have therefore modified this entire section of text to make it clearer.

“Marécal et al. (2012) showed that significant amounts of Br₂ were only released into the gas phase from R7 in cloud droplets when their idealized simulation was initialized with very high CHBr₃ (40 pptv) in the BL. In the more realistic case where boundary layer CHBr₃ was ~1.6 pptv, the formation and release of Br₂ via R7 was very limited, which is consistent with our findings here as shown in Fig. 15. Following the causal link we identify between low ozone and resulting low HBr and HOBr, these similar results for Br₂ could be explained by the similarly low background O₃ simulated over the two regions (Borneo and Darwin) in both studies. In Marécal et al. (2012) the background O₃ was 14 ppbv at 2 km as compared to the 10-15 ppbv range simulated over the inner domain at 2 km altitude in this study.”

19. Section 4. How do the results from this paper fit in the context of findings from the CONTRAST field campaign? I am not sure it is possible to have a thorough discussion, but noticed that Chen et al. (2016) JGR discuss convective transport (or not) of BrO and HOBr + Br₂.

We thank the reviewer for highlighting this relevant paper. This study offers some qualitative support for the hypothesis linking both the low simulated BrO and HOBr levels and also the limited production of Br₂ to the low background tropospheric ozone levels. Indeed, they only observed elevated BrO and HOBr+Br₂ in biomass plumes where ozone was significantly elevated and for the remainder of the time the observed levels of BrO and HOBr+Br₂ in the troposphere were below the limit of detection. We have therefore added some text in Sect 4.3.4 to discuss this.

“Indeed, there may be some observational support for this in data collected during the CONTRAST field campaign. Chen et al. (2016) showed that the observed levels of BrO and the sum of HOBr+Br₂ were both below the limit of detection (0.6-1.3 pptv and 1.5-3.5 pptv, respectively) in background tropical tropospheric air where ozone levels were relatively low (< 50 ppbv). They found that BrO and HOBr+Br₂ were only above the limit of detection in biomass burning plumes where ozone levels were significantly elevated (> 50 ppbv). Their findings are at least consistent with our expectations of a link between ozone and the bromine speciation between HBr, BrO, HOBr, and Br₂.”

20. Section 5. Can results from global model studies (e.g. from TOMCAT, or other CCM1 models) help identify other tropical regions that should be studied?

The work of Hossaini et al. (2016) highlights the Indian sub-continent and South-East Asia during the summer monsoon as two regions for importance for vertical transport of bromoform linked to deep convection. Their work shows overall lower levels of bromoform being transported to the upper troposphere, but the background tropospheric composition is likely very different due to the proximity to large sources of anthropogenic pollutants and ozone precursors. We have now highlighted this region in section 5 with:

“Hossaini et al. (2016) highlight the Indian sub-continent and southeast Asia as another region that is potentially of importance for transport of bromoform and its PGs to the upper troposphere within deep convection. They predict it to make a more minor contribution to the vertical transport of bromoform compared to the maritime continent, but the background tropospheric conditions are very different from the present study due to its proximity to large pollutant and ozone precursor sources, and its contrasting conditions could make it an interesting case study.”

Line 60, I suggest starting the sentence as, “Bromoform (CHBr₃), with 3 Br atoms”

We have modified this sentence as suggested.

Line 505, Could you make the negative ion sign more obvious. Perhaps via a symbol or equation editor.

Thank you. We have tried to fix this by increasing the font size.

Line 589, Suggest: Fuhlbrugge et al. (2016) who showed that

Thank you. We have changed the text to correct this.

4. I recommend that the Appendix be placed in the supplement.

We have moved the appendix to Supplement S2.

In Table 1, is the Hx for Br₂ correct? A positive value is given for aH yet Hx is less than H₂₉₈.

Correct. This was an error. We have modified Hx to the correct value for Br₂ in this case, which is 0.97.

Is the vertical cross section in Figure 8 in a different location than the other vertical cross sections?

This is the same location as shown in Figure 9c etc, which corresponds to Mod_Conv 3 located at 5.4°N. Figure 8c corresponds to the same time (9 UTC) as in Figure 9c etc.