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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Cloud-scale modelling of the impact of deep convection on the fate of oceanic bromoform in the troposphere: a case study over the west coast of Borneo" *by* Paul D. Hamer et al.

Anonymous Referee #1

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The paper by Hamer et al. discusses a case study of convective uplift of bromoform over the west coast of Borneo. The authors first analyze the spatial and temporal evolution of two convective systems based on satellite cloud top temperatures, which have been probed in research flights during the SHIVA campaign. The same region is then simulated with the C-CATT-BRAMS atmospheric model and three similar convective systems are identified. The simulated CHBr3 is then compared in a statistical way with measurements of CHBr3 in observed convective systems. Further, convective systems are analyzed in cross sections regarding their uplift of CHBr3 and

its product gases.

This paper addresses an important topic which is certainly within the scope of ACP. However, the manuscript misses to make one consistent work out of several interesting studies. The authors should certainly better motivate each part of their study to illustrate how these different parts contribute to the scientific questions addressed by this paper. In addition, I have identified several major issues and specific comments that need to be addressed before resubmission. I recommend resubmission of this manuscript after major revisions have been made based on the listed issues and comments below.

Major issues:

- The connection between measurements and model study is not clear to me. Measurements from the SHIVA campaign are only used marginally and mostly are used to motivate to simulate the west coast of Borneo in the model study.
- One of the connections between measurements and model is the statistical comparison of measured and simulated CHBr3 mixing ratios at BL, convective and nonconvective UT. Unfortunately, the usage of statistical quantities in comparison with measurements seems to be very arbitrary. The authors should first introduce each statistical quantity and tell the reader what they want to show by examining this specific statistical measure. Then they can proceed to discuss each quantity.
- I was quite disappointed that the only comparison to aircraft measurements was in a statistical way. As indicated by Fig. 2, there are CHBr3 measurements from transects through the Obs_Conv1 convective system. The authors are right that a lat/lon-based comparison between measurement and model would be not reasonable due to the local displacement of the convective systems. But why do they

not show CHBr3 transects through the simulated Mod_Conv3 convective system on similar altitudes as measured? For a comparison, measured and simulated transects could be aligned in the center of the convective system. In my opinion, such a comparison would much better demonstrate a possible agreement between model and measurement compared to only the statistical comparison.

- Throughout the manuscript, but in particular in Section 4.3., figures are not well introduced in the text. Some parts of figures are not even mentioned at all in the manuscript. The authors should either remove these parts from the figures or introduce them in the text. See also in the specific comments.
- The discussion of the PGs is not well motivated. What is the aim of this very lengthy discussion? Do the authors want to explain mixing ratios of CHBr3 PGs in the convective systems, in the convective outflow or in the tropospheric background? With the given plots it is not demonstrated that PGs increase in the upper troposphere due to convective uplift of CHBr3 or PGs. How can the enhanced values of PGs be discriminated from enhanced values of PGs due to background CHBr3 that has been in the upper troposphere without convection? Are PGs transported upwards by convection or are they rather formed in the UT from transported or background CHBr3?
- All discussions about upward transport are limited to transport to the upper troposphere. It is mentioned by the authors, that CHBr3 and PGs are of high relevance in the stratosphere, but it is not even mentioned if brominated air masses reach the stratosphere through the convective systems (in fact, not even the tropopause is marked in any plot). Why is this possibility neglected in the study?

Specific comments:

- 60: "CHBr3, with 3 Br atoms per molecule, has the largest emissions among the different brominated VSLS.": Please quote a suitable reference for this statement.

- 82: "Convective transport and the associated chemistry and washout of all bromine containing species (Bry) cannot be simulated in detail with global 3-dimensional models because of their coarse resolution, and because of the complexity of the chemical processes (e.g. Hossaini et al., 2010).": Why are the authors mixing the two topics here in one sentence? The following sentences are only about convection, not about chemistry and washout, so I don't see any need to introduce this topic already here. In addition, I would talk about "current state-of-the-art global 3-dimensional models" not being able to resolve all convective events.
- 87: "Regarding chemical processes and their interactions with liquid and ice hydrometeors, global models have made progress (Hossaini et al. 2012, Aschmann and Sinnhuber, 2013, Liang et al. 2014), but they still need to compromise between complexity and computing resources.": This sentence is very vague. What are the progresses that have been made by the models? What are the relevant chemical processes and interactions with liquid and ice hydrometeors for CHBr3? Some more detail is missing here.
- 82-93: The whole paragraph should be restructured. After reading it multiple times it became clear to me that the authors are talking about chemical processes during the convective uplift process. This should be mentioned in the beginning of the paragraph, not in the last sentence. In addition: What are the most important reactions of CHBr3 that need modelling in kilometer-scale resolution?
- 97: "... within the optimised mechanism." I'm not a native speaker, but "within" sounds wrong to me. Maybe better use "based on"? But I certainly may be wrong here.
- 129 (and following occurrences): I find it hard to remember which of the convective systems was called "Obs_Conv1" and "Obs_Conv2". I don't see any reason why the authors would need to number these systems. For me it

would be much easier to call them either by their colors (e.g.: "Obs_Conv_blue" and "Obs_Conv_pink") or maybe according to their geographical positions (e.g. "Obs_Conv_northeast" and "Obs_Conv_southwest"). The same comment applies to the "Mod_Conv*" named convective systems later in the manuscript. In addition, the "blue box" in Fig. 1 looks not really blue to me. Maybe the authors could increase brightness and saturation of this color?

- 137 following: The authors could help the reader by either mentioning the panel character in Fig. 1 in the text (e.g.: "Obs_Conv1 was already well developed at 05 UTC (13h local time: 13 LT; Fig. 1a) ..." or by writing the UTC times directly in the panels of Fig. 1. Of course, it is stated in the caption, but for me it took some while to find the corresponding panel from the description in the text.

- Fig. 1: The colorbar used for the brightness temperature includes several maxima and minima in brightness, which distorts the perception of the plot. It is, for example, very difficult to see if the brightness temperature of the blue box convective system increases from panel (f) to (g). Please use a different colormap. Colormaps used for Fig. 8 and following are much better. In addition: A grid line for 117°E longitude is missing in the map.

- 144 following and Fig. 2: I'm not really sure what I should see in Fig. 2. The flight path was very complicated and even though the authors tried to mark certain points with time stamps it is not possible to reconstruct the flight path. If the purpose of Fig. 2 should be to prove that measurements have been taken inside Obs_Conv1 and Obs_Conv2, I would suggest to repeat the 9 UTC brightness temperature measurements of Fig. 1 in the background of Fig. 2. Or maybe it is even possible to integrate the flight path (using different colors) in Fig. 1?

- 152 following: "This scenario was confirmed ... CHBr3 measurements were performed by the GHOST ..." The last sentence of this paragraph seems out of

place and gives little information that could have been included in the previous sentence. Also, GHOST has not been defined yet.

- 159: The authors state that "This system is capable of resolving meteorological processes ...", but later it is described that it "includes various physical parameterizations to simulate sub-grid scale meteorological processes ...". The important question for this manuscript is: Is deep convection resolved by the model or parameterized?

- 167: Please define "MODIS NDVI"

- 190: Please define "Fast-TUV"

- 198: "For the model to simulate ..." This sentence sounds strange. Maybe: "Several important changes have been applied to the model to simulate chemical and physical processes associated with CHBr3 degradation chemistry and transport."

- Fig. 4: The authors could help the reader to find Borneo on the coarser map by marking the detailed map boundaries that have been used for Figures 1 2.

- 280 "Therefore, Fig. 5 is mainly used here to show the general temporal and spatial development of the simulated convective systems but does not provide a precise measure of the cloud top height and spread of the anvil.": If this is the purpose of Fig. 5, I do not understand why the authors chose to present cloud top altitudes and not an approximate conversion to brightness temperature. In the given representation of Fig. 5, it is almost impossible to compare to the temporal and spatial development of the measured convective systems in Fig. 1. I would strongly recommend to change Fig. 5 to brightness temperatures instead of cloud top altitudes.

- 283: Same comment regarding the names "Mod_Conv*" as for line 129.

- 299: "Table 2 shows a general agreement on times and altitudes between the observations and the model." This statement is not true for the timing of the convective systems. The simulated convective systems either are started later (8 UTC in the table, but 9 UTC in Fig. 5 compared to 5 and 7 UTC in the measurements) or dissipate earlier (8 UTC compared to 11 UTC in the measurements).
- 308: "the duration of the system of several hours and decay during early evening.": Same here. The duration of the system is always simulated considerably shorter than measured.
- 322: I would call this section differently - it is the part where the model finally is compared the measurements - so I would mention the comparison to the measurements in the title of the section.
- Table 3: Please give units if applicable (I guess it is pptv for all [X] quantities). Also, fraction f is given with a kind of uncertainty (+-...) but in contrast to the [X] quantities, it is not stated what kind of uncertainty is presented.
- 344: "However, this high fraction f is consistent with the average value calculated from all SHIVA aircraft data ...": The authors should introduce the fraction f better. Is it expected to be the same factor f for all convective systems or is it expected to vary between individual convective systems? In the latter case, it would mean that Mod_Conv2 is not comparable to Obs_Conv1 or Obs_Conv2 in terms of the fraction f. It is also worth noting that Mod_Conv2 has a higher uncertainty and agrees with the fraction f of Obs_Conv1 and Obs_Conv2 within the combined uncertainties.
- Figure 7: Please add a legend explaining the colors. It would be also helpful to use the same colors for Mod_Conv* regions as in Fig. 5
- 353 and following: From column [X]BL in Tab. 3, I would say that the model results agree to the measurements within their uncertainties. I don't see the

rationale of discussing single differences in percentage numbers here. For the whole following paragraph it is not clear to me why there are so many different statistical measures used. I would recommend to restructure this paragraph and first explain which statistical measure is used for what purpose. In the current state, the authors jump from a discussion of mean values (without mentioning the 1 sigma errors) to the median with 25 and 75 percentiles. What do I learn from these numbers? The same issue continues for the following paragraphs.

- 366: "If we consider the higher spatial resolution of our simulations and the smaller domain considered for the statistics compared to TOMCAT, these remaining differences appear consistent with one another.": I don't understand this sentence. Please give some context.
- 369 following: This paragraph and the following (starting at line 378) are very similar to the previous paragraph and discuss the differences in background CHBr3 in the UT. Some arguments are repeated in these paragraphs, some are new. Please restructure these three paragraphs to one without repetitions. Instead, a discussion of UTconv CHBr3 is missing completely here. Also median and 25 and 75 percentile information from Fig. 7 is not used here at all.
- 389: "We selected Mod_Conv3 since it corresponds mostly close in space to Obs_Conv1.": This seems a good choice, but unfortunately, the authors do not compare this simulated CHBr3 to observations.
- 393: "...naturally highest closest to the point of convective detrainment..." Please check the formulation. Maybe "... naturally highest, close to the time of convective detrainment ..."?
- Figure 8 and following figures: Please label at least one axis per column and row.
- Figure 9: A short notice would be helpful that Fig. 9c is the same as Fig. 8c scaled by the number of bromine atoms (3). It would be also helpful to repeat the

black and white lines from the first row of Fig. 9 to all other rows and the following figures to guide the eye in a comparison. In addition, these black and white lines would help to identify regions of convective outflow and tropospheric background. Also a tropopause would be helpful for all these kind of plots.

- 438: Missing ")" after "(i)"
- 452: Please define the gases that are summarized as "bromo-carbonyls" and "bromo-methyl peroxides" or use the same names for these groups as in the introduction.
- 453: "CHBr3 is insoluble relative to its PGs" -> "CHBr3 is less soluble than its PGs"
- 469: "These compounds contribute 86
- Figure 14 d-f: The representation of HOBr as percentage of total inorganic Br mixing ratio is dangerous here, because this plot suggests that HOBr has a large contribution to the convective system by showing relative contributions up to 100
- Section 4.3.3: There are plots for insoluble organic bromine compounds in figures 11 and 12, but these are not even mentioned in this section.
- 470 and following: In this paragraph, the "behaviour of inorganic bromine" is discussed. Earlier in this section inorganic bromine was introduced as a large number of species, but here only HBr and HOBr are mentioned. This selective discussion of only two gases needs to be motivated.
- 479: "A key finding is that inorganic bromine dominates the PG budget within the troposphere, yet despite this the inorganic PGs are almost entirely removed during convective transport by washout due to their solubility." This is a broad statement based on a case study. I think the authors should limit this statement to their case study and not leave it in a general sense.

- 480: "We here argue that the regional tropospheric composition present in our simulations is the underlying cause of this prevalence of HBr and in turn the washout of inorganic PGs that results from this.": I find this sentence very unclear and don't understand what the authors try to say here. Please rephrase. This makes it also very hard to understand the motivation for the whole Section
- 4.3.4. Why is so much discussion devoted to the chemical processes?
- 523 following: The comparison to the Marécal et al. (2012) study is very interesting but comes very abrupt here. The authors should consider giving this comparison its own subsection.
- 559: "First, it could be difficult to ..." –> "First, it is difficult to ..."
- 561: "Furthermore, other tropical regions could have vastly different CHBr3 emissions, and in the case of much higher emissions, as was explored in Marécal et al., (2012), we could expect a larger role for Br2 formation." Please check the grammar of this sentence. In addition: What exactly is expected to have a larger role for Br2 formation?
- 570: "Despite the difference in simulated CHBr3 mixing ratios ...": Differences to what?
- 573: "Indeed, our results show consistent CHBr3 mixing ratios compared to the simulations of Hossaini et al. (2013) for the November 19th flight.": To my understanding this consistency between two models using the same emission scenario only proves that both models work properly in terms of chemistry and transport, but it does not prove that the emission scenario is useful, as it is intended by the authors here.
- 593: "Most of the bromine (>85

- 595: Missing subscript in CBr2O
- 598: "Overall, we conclude that organic PGs are more important than inorganic PGs for the vertical transport of bromine within the convective columns for the conditions that we study here." This is not true, because it was stated earlier in this paragraph that most of bromine that was convectively transported was in the form of CHBr3. Maybe the authors want to limit their statement to the vertical transport of PGs.
- 606: "... more important role of the inorganic PGs for the vertical transport of bromine.": The authors have not shown that the enhancements of HBr in the upper troposphere are due to convective transport.
- 608: "Overall, these conclusions are valid in all parts of the convective system except for where the anvil detrains into residual convective outflow in the UT.": Such a statement is not covered by the findings discussed in the main part of this paper.
- 612: In my opinion, section 7 is very short and could be attached to section 6.

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