acp-2013-585 – author response to reviewer 1

The paper "Estimates of tropical bromoform emissions using an inversion method" is a novel approach to derive emission estimates from a short-lived compound, emitted from the oceans in significant quantities, but with large uncertainty in magnitude and distribution of the emissions. The authors present back-trajectory calculations, dilution matrixes and several possible solutions for an inversion method for new emission estimates. It is well justified to use this approach, and discuss its benefits and limitations. The paper is well written and the modeling appears profound. The authors discuss their method and uncertainties and possible improvements mostly in detail. Some terminology which the authors have used or created should be explained a little more precisely and the inversion method should also be described a little more in detail. The authors shall refine some statements and explain at some places in the manuscript a little bit more of the background they used to obtain the results, which should be discussed a little more. It did not become clear why the authors did some experiments – appear redundant – or shall be better explained. The differences in the solutions (25) for one study case where not shown and the reader wonders how large they are. The question is if the differences can be somehow shown? In other words how robust is such a mean emission distribution estimate for one case study? The authors should discuss if this new method improves the information about the "real" regional emission distribution of CHBr3 and add a little discussion about the possibility to pin down emission sites for CHBr3 with this method. The abstract is saying … obtains detailed estimates of the distribution, which is not followed in the final discussion. The discussion is missing a little bit of concluding highlights of this new approach and study - which are somehow buried in the text- under possible improvements. Thus I suggest to highlight the highlights and point out what is new and brilliant about this approach, before suggesting the possible improvements, which may even strengthen the paper. All in all it is a good paper already which needs minor revisions in the text and some details, which are addressed in the specific comments.

We thank the reviewer for the very positive comments. We are also grateful to the reviewer for highlighting areas where out description of methodological details might benefit from some clarification. We have tried hard to thoroughly address the specific comments (below).

We have been asked to highlight 'what is new and brilliant about this approach', and as a first step, suggest revising part of the abstract. We will now write: "In this study we aim to reduce this uncertainty by combining the first multi-annual set of CHBr3 measurements from this region, and an inversion process, to investigate systematically the distribution and magnitude of CHBr3 emissions. The novelty of our approach lies in the application of the inversion method to CHBr3." To avoid repetition, we will also rephrase a subsequent part of the abstract as follows: "We then obtain detailed estimates of CHBr3 emissions within this area, which appear to be relatively insensitive to the assumptions inherent in the inversion process."

Next, we will add sentences in the introduction (at page 20465, line 26) that highlight the fact that CHBr3 emissions have never before been estimated using an inversion method: "Inverse methods, in which both the magnitude and distribution of emissions are varied to provide an optimal match with observations, can be considered an extension of the top-down method. Our use of an inversion method in this study represents a novel approach to estimating CHBr3 emissions." We will also slightly re-write the first few sentences of the final paragraph of the introduction.

We also plan to rewrite the first few sentences of section 6 as follows: "In this study we have described a new method to quantify CHBr3 emissions. For the first time, an inversion process has been used to investigate systematically the distribution and magnitude of CHBr3 emissions. This process is informed by new long-term, high-frequency observations from Borneo, which are the first of their kind in the poorly sampled and convectively active Maritime Continent." We will also change "means" to "has the important implication that" on line 13 of page 20483.

We have then been asked to say a little more about the geographical details (and the utility) of the emission distributions we have calculated. We have compared our emission estimates (e.g. from experiment D) with two commonly available, possible proxies that have been covered in the literature (see also our reply to reviewer 2). However, our estimated CHBr3 emissions bear little resemblance to the distribution of either chlorophyll-a, or of ocean depth. Our study therefore echoes previous findings: no simple relationship that would enable a straightforward physical parameterisation of CHBr3 emissions appears to exist. To cover this point we will add sentences in section 5.2 - where we also intend to add a new discussion of how representative of the tropics (in terms of these two possible proxies) our study area is - and to section 6.

Specific comments:

Page 20466 Line7: include Ziska et al., 2013 (as bottom-up approach) in the references with Liang and Ordonez

Yes, Ziska et al., ACP, 2013 suggest CH2Br2 emissions at a similar level to the other two cited papers. We will cite this third paper here.

Page 20467 Line 19: why did you choose 3.51 ppt as threshold?

We assume the reviewer is concerned with the apparent precision of our threshold. This choice is important for our inversion calculations, and while we accept it is somewhat arbitrary, we already argue (on page 20479, line 20 onwards) that this choice is reasonable, and that our results do not depend upon it. To aid clarity we suggest slightly rephrasing this sentence as follows: "The CHBr3 mixing ratio at Tawau, on the coast, is occasionally many tens of ppt, with ~10% of the observations above ~3.5 ppt (a threshold marked by the red line in figure 1, and discussed with respect to our inversion calculations in section 5.1)."

Page 20469 Line 19-22: The explanation of the generation of the "dilution matrix" appears insufficient. Since the generation of the matrix is a crucial step in the method, please describe it in a little more detail (according to Manning 2011).

Reviewer 3 makes a similar comment, and we are happy to add more detail and clarification. Suggested text (as a new paragraph) to replace the sentence currently occupying lines 19-22: "In more detail, each trajectory is assigned an arbitrary mass, and the density of trajectories in each grid cell, integrated over the 12 day travel time (units are g s m-3), is recorded. This density is then divided by the total mass of trajectories released (g), and multiplied by the area of the grid cell (m2). Physically, the resulting quantity (which has units of s m-1) can be considered the mean residence time in a particular grid cell, for all trajectories started in a particular 3h period, divided by the cell depth (100 m). Importantly, this quantity is the multiplicative factor by which emissions in a particular grid cell are diluted by the time they have travelled to a measurement location (see equation 1, and Manning et al., 2011). So, a low dilution value means that few trajectories reaching Bukit Atur or Tawau have travelled through the boundary layer of a particular grid cell, and therefore that the impact of emissions from that grid cell on a measurement site will be correspondingly small. A grid of dilution values exists for the batch of trajectories started in each 3 h period. We will refer to the full three-dimensional (longitude-latitude-time) grid of dilution values as the 'dilution matrix'."

For consistency with the above, we also propose to adjust the start of the subsequent sentence covering lines 22-26 so that it reads: "We approximate the effect of photochemical loss of CHBr3 by allowing the mass associated with each trajectory to decay with an e-folding time of 15 days …"

Page 20470 Line 6: Here also you naturally assume, that the reader can transform easily between dilution values and the number of trajectories in a certain grid box. Explain the connection between the number of trajectories and the dilution value –at least in one sentence or above.

We hope the additional explanation outlined above will solve this problem.

Page 20470 Line 10: What does less dilution information mean?- You have a factor- why is there less information, about what? Please revise wording or sentence. Actually, I do not understand what you want to say here with this sentence?- Possible erase or move, or revise?

Again, we hope the above explanation is helpful. We agree that the wording can be improved and suggest concluding this sentence with 'where the dilution values are smaller'.

Page 20471 Line 8: Aren't they also uniformly distributed?- constant in space?- or do you put different emissions in each grid cell?

We hope that adding the following text after equation 1 will be helpful: "From equation 1 we obtain a spatially varying emission field. In solving this equation we assume the emissions are constant in time."

Page 20471 Line 17: the content after the ", " is redundant since natural, or do you want to say something different ..please rephrase then.

We agree that we are perhaps stating the obvious, but think it is still worth pointing this out.

Page 20471 Line 20: Does this help for a variable compound such as CHBr3, since then you also group the emissions to a non existing mean and therewith loose information?

Grouping the grid cells does lead to a loss of spatial information. The argument for taking this step is that considered alone, the more distant individual grid cells are crossed only rarely by a small numbers of trajectories. The inversion method will then have very little information about that individual grid cell with which to determine an emission. Grouping the grid cells increases the quantity of information available to the inversion method (but of course at the cost of spatial resolution).

Page 20472 Line 4: concentration in what? atmosphere, grid box, receptor site?

Following the suggestion of reviewer 2, we propose changing this text to: "… be used to derive a simulated concentration at both measurement sites …"

Page 20474 Line 10ff: could you please include, which observations (3h means?) and modeled concentrations you use? Which number is behind the n?

We suggest changing the text preceding equation 2 to: "We use the normalised mean square error (NMSE, Eq. 2) as a cost function to judge the strength of the agreement between the simulated concentrations (m) and the three hourly average observations (o), and it is this error that the optimisation process will seek to minimise. In Eq. 2, n is the sum of the number of observations at both measurement locations (2350+2726=5076, see figure 1)."

Page 20474 Line 21: ..it is known that the emissions at coastlines are more significant..and you apply this later..?! how does this fit to your statement?

While it is known that strong CHBr3 sources (typically seaweed) are often found in coastal regions, we would argue that the detailed distribution of CHBr3 sources (or seaweeds, or other possible CHBr3 sources) is not well known. For example, we do not want to assume that emissions along all coastlines are uniformly strong. It is also not clear how much of a regional impact the strong coastal sources have (i.e. how important are emissions from seaweed relative to emissions from the open ocean).

Page 20475 Line 23: are the 25 times independent of each other or did you make them subsequently? please clarify

We propose changing the text to: "… is independently repeated 25 times …"

Page 20478 Line 14: I still do not completely understand, how the model calculates the emissions. Please explain a little more detail with equation 2. Individual solutions must yield very differently distributed emissions for each of the considered cases? Is it worth showing this?

This is an important part of the paper, so we hope that the following additions will clarify the process followed to find an emission distribution.

First, we will refer to Eq. 1 in the sentence starting on line 5 on page 20474; we suggest changing to: "… the modelled time series, calculated using Eq. 1, and the observed time series."

We will slightly rewrite the first paragraph on page 20471, so that it reads as follows: "The inversion method employed here has been described by Manning et al. (2003, 2011), and is now known as the Inversion Technique for Emission Modelling (InTEM). Using an optimisation process, the aim is to solve equation 1, and thereby locate the emission map, that can be multiplied by a NAME dilution matrix, to give modelled concentrations at a measurement site that most closely resemble the observed concentrations (converted from volume mixing ratios using UM meteorological data)."

We then plan to add the following text, on page 20471 immediately after equation 1, to improve our explanation of the emission calculation: "This calculation is conducted for each individual (source) grid cell: the emission in that grid cell, multiplied by the local value of the dilution matrix derived from the trajectory calculations, gives the contribution from those emissions to the simulated CHBr3 concentration at a measurement location (receptor). The sum over all source grid cell contributions gives the total simulated CHBr3 concentration at one particular time."

In addition, we will insert the following text on page 20475, line 8: "In brief, the method investigates a solution space of possible emission configurations. It iteratively searches the solution space to find the emission map that, after transformation to a modelled concentration using the dilution matrix, most closely matches (as measured by Eq. 2) the observed time-series."

We also hope that our improved description of the dilution matrix will be useful.

We agree (as does reviewer 2) that it would be helpful to provide more information about the variability within the 25 individual solutions to each inversion. As an aid, we would like to include a new figure (Fig 7) that shows the individual solutions containing the minimum and the maximum estimated emission in experiment A. We will place more emphasis on the fact that the range between the lowest and highest emission calculated for the fine grid cells is relatively small (1.2-1.4 Gg CHBr3 yr-1). We suggest the following text be inserted after the sentence that finishes on page 20478, line 16: "The individual solutions to experiment A containing the lowest and highest emission are shown in figure 7, plots a and b; while the grid cell by grid cell details of the distribution of emissions in these two solutions are somewhat different, the main

features are consistent, with emissions found largely in the oceanic parts of the map, and much smaller over land."

We will also fix a typo in table 2: the mean and range of emissions for Exp A should be 1.3 (1.2–1.4) rather than 1.2 (1.3–1.4).

Page 20478 Line 16: How did you model these concentrations ? forward trajectories?? And is it not expected that they yield the same concentration, since they use the same input as the inversion?

Reviewer 3 also requested that this calculation be clarified. The concentrations were calculated using Eq. 1 (i.e. from the dilution matrix and from the estimated emission). We hope that the additions we suggest above will have clarified this point, but will explicitly mention Eq. 1 here too: "Using the emissions from experiment A and the dilution matrix we now solve equation 1 to obtain simulated CHBr3 concentrations at Bukit Atur."

Page 20480 Line 16 to 22: I do not understand also in connection with table three, what you did here and what you like to show here. E.g. What is your choice of grid cells? Can you clarify this also in the text, what are the consequences of this investigation?

The motivation for including table 3 was to support our choice of 'fine' grid (i.e. including grid cells of both 1 by 1 and 2 by 2). Given that reviewer 3 also did not find it particularly helpful, we suggest removing this paragraph, table 3 and the one other mention of it in the text. This should not affect any other sentences.

Page 20482 Line 4 to 8: Why do you need to consider fine versus 4x4 and for what region? The whole issue with the choice of the size of grid cells for what region and what you want to show here shall be clarified throughout the text.

We hope that by slightly re-writing these sentences our method will be clearer. We propose: "We can also consider the extent to which our definition of 'fine' grid cells impacts upon our extrapolation. If 'fine' were defined with a maximum grid size of 1 by 1 then extrapolation gives a total tropical emission of 180 Gg CHBr3 yr-1. Conversely, if the maximum grid cell size were 4 by 4 the total tropical emission after extrapolation is 240 Gg CHBr3 yr-1. These values are summarised in Table 1. They also fall within the range given by experiments E and F, and show that our choice of 'fine' grid has had a relatively small impact on our extrapolated emission magnitude."

Page 20483 Line 1: May be it would at least be nice to see what difference you got for the coastal area?

We suggest providing this extra detail after the final sentence in this section which ends on page 20483, line 2: "To illustrate, in the case of Experiment D, our original definition of coast, based on the UM land/sea mask, led to 25% of emissions in coastal areas, and 67% in open ocean areas (see Table 2). Using this new definition the equivalent percentages are 16% and 72%."

Page 20483 Line 16 and 17: This is a very coarse statement- and there are planktonic sources in the open ocean- while algae are assumed with coastal macro algae and Zulu Sea is not necessarily open ocean. And didn't we expect coastal region to deliver more? Please refine.

We had hoped that 'algal' would be read as both macro- and microalgae. We intend to make this explicit in the sentence.

Page 20484 Line 1 to 8: Didn't Hossaini also show that the low Ziska emissions for CHBr3 matched best in the tropics?

Agreed – Hossaini et al. reported that, of the inventories considered, the Ziska emissions appeared most consistent with the tropical data considered. We will mention this.

Page 20484 Line 11 to 12: How are the emissions in the receptor grid box resolved in your inversion model? This must cause a large error if they are not included? Please discuss, how relevant this is for the short-lived compound.

The two 'receptor' grid cells are treated the same as all others, and emissions are assumed uniform within these cells. This is clearly not an accurate reflection of reality, and is why we attempted to remove the influence of very local emissions at the coastal site by removing the highest observed mixing ratios from our calculations.

Page 20485 Line 20: This could be elaborated a bit more – in the chapters before and here please try to specify this statement.

We suggest appending the following text: "We have made progress in this direction by starting to make measurements of CHBr3 and other halocarbons at sites near Darwin, in Northern Australia, and in Bachok, on the East coast of Peninsular Malaysia."

Numbers in Figure 4 and Table 1 for accumulated emission form the different resolution grid cells don't match , % in Figure 4 for 1x1 is 44 %, while in Table 1 it is only 2% .(or is Egrid not for all 1x1 cells?, then please rephrase clearer. Wouldn't it be sufficient to define " fine grid" cells always the same way..not: as in the black line, or red and orange, just e.g. fine (1x1 plus 2x2)? Too many definitions under the pictures for the same thing

I'm afraid we are not entirely sure what the reviewer is asking for here, but hope that the following explanation will be helpful:

The numbers in Fig 4 refer to a cumulative contribution to a simulated concentration (see also our subsequent response) under an idealised situation (i.e. not an inversion experiment) in which uniform emissions are

assumed across the entire solution domain (the full grid in Fig 3). We describe this in more detail on page 20472.

Table 1 summarises results from inversion experiment D, and Egrid refers to the total emission from grid cells of a particular maximum size.

Our base assumption is that 'fine' cells are or size 1 by 1 and 2 by 2. However, we have endeavoured to show (in Table 1) that our conclusions about CHBr3 emissions do not depend on this assumption and that we would reach similar conclusions if 'fine' was defined as only the 1 by 1 cells, or if 'fine' was defined as the 1 by 1, 2 by 2 and 4 by 4 cells.

We feel that we are introducing a source of confusion by referring to table 1 in the same paragraph we introduce figure 4. Accordingly we will remove the reference to table 1 on page 20472, line 9. We will also include a little more information about table 1 within the parentheses on line 24 of page 20472: "(the areas covered by these different grids, along with estimates of emissions in one of the subsequent inversion experiments, are given in Table 1)".

Figure 4: what is accumulative emission sensitivity?

For consistency with the text on page 20472 and the caption, we suggest re-labelling the axis as 'cumulative contribution to simulated concentration'.

Table 3: I do not see what this table is aiming at- in addition I do not understand what you mean with maximum grid size and total emission from grid cells (is it all cells or per cell?) area –averaged flux per cell? Can you choose a different unit to increase the numbers?

As suggested earlier, we intend to remove this table.