

We thank both referees for their constructive comments, the main points of which we summarise as follows:

1. The data is interesting; however, there is more data which could be shown. Also, the analysis of the data could be more quantitative/substantive.
2. The modelling analysis lacks detail; the use of the two models is confusing. The interpretation of the observations and model results in terms of global emissions is uncritical.

We accept these comments and have addressed them in our response. We believe that we now have a much stronger paper in consequence.

1. First, we have stressed that one motivation for this study is the need to show that the instruments we have constructed are capable of independent operation, allowing consistent measurements. Demonstrating consistent side-by-side operation is an important prerequisite for our future measurement plans and one aim of this paper.

This paper was written for the OP3 special issue. We believe the issue of spatial variation discussed here is of general applicability to short lived species measured in this (and other campaigns). Although we now have much more data, this is not completely analysed yet. So, we confine our attention to measurements during OP3.

We have added further analysis of the data collected. The revised paper will include a new figure showing pdfs of the Danum data (showing that the two instruments do indeed measure essentially the same concentrations) and of data when the instruments were deployed simultaneously at Danum and Kunak. In this case the pdfs of the measurements are quite different.

2. We have rewritten section 4, “Modelling and data interpretation.”. We begin with a much clearer statement concerning the use of the models. NAME is used primarily to study local variability and the differences between Danum and Kunak; to understand (and provide an estimate of) regional emissions. We present more details of our NAME calculations including figures to show the contribution to modelled tracer arising from different locations. pTOMCAT is used to put these emissions in a global context. Again, we have added discussion of some new calculations. We have added a much more detailed discussion about the extent to which local measurements can (or cannot) be used to make global estimates. There is more emphasis on uncertainty (which we had mentioned in our original manuscript but, from the referee comments, this was clearly insufficient).

We have written a couple of paragraphs to discuss issues related to model uncertainty, including model resolution and the treatment of the boundary layer.

More details of the models’ formulation, especially for pTOMCAT, are given in section 2.2, as requested.

We turn now to the specific referee comments.

Ref 1.

General comments

This paper present new measurements of bromoform in the tropics, a key source region

for short lived halogen gases which may be transported to the stratosphere and contribute to ozone depletion. The authors find much higher concentrations at the coast than about 50km inland, which they could not explain at the resolution of the models used, nor were the absolute levels consistent with current emission scenarios. The manuscript represents an important data set, and is in scope for ACP, however I do not find that it reaches substantive or quantitative conclusions. For example, what are the general implications for bromoform source strengths? It is not made clear whether the mismatch is really due an overestimation of sources (by Warwick et al and others), or rather to a combination of coarse model resolution and local sources. If the latter – it would be good to show this using a higher resolution model, and more information on the local macroalgal densities.

Our revised document gives a more quantitative and substantive discussion about the implication of our observations and our modelling studies for the determination of source strengths. We have also added much more discussion concerning the difficulties (eg resolution) in determining sources.

Further, the model results are not described accurately. For example, in the conclusions the authors state “Both models show that, despite a lifetime of about two weeks, substantial gradients between the coast and inland can be expected for bromoform, with the coastal measurement variability being dominated by local emissions.”. This is not the case – Fig 5 for example shows very little difference between the model predictions for the 2 sites. Further, there is no systematic analysis of variability to back up the latter statement.

The section on modelling has been changed substantially and the philosophy of using two different models is set out much more clearly. Thus, it is NAME that is used mainly to study the gradient between coast and inland. Here, we add to our discussion by showing NAME-modeled tracer pdfs. The discussion of Figure 5 is changed and we also show pdfs from the pTOMCAT results.

The final point is that the derived global source strength for bromoform is clearly subject to a high level of uncertainty, which is not made clear.

We did mention uncertainty but this is now investigated, and emphasized, much more strongly.

Major comments:

p14978 and Fig 4.

Ln 20 “The results in Fig. 4 show some similarities to Fig. 1. First, the concentrations at Danum are usually less than at Kunak, sometimes by a factor of two or more.”

This statement is rather confusing. The results in Fig. 4 seem to show no resemblance to those of Fig 1, in particular there appears to be little difference in Fig 4 between the Danum and Kunak model results. I think the caption means that the Danum results are scaled to the Danum measurements and the Kunak model results to the Kunak measurements, but this should be rewritten to make this much clearer – or even better – simply leave out the scaling altogether.

This has been changed. The NAME results at Danum were, in fact, normalized such that the mean modeled tracer concentration matched the observations. The same normalization factor was then applied to Kunak.

Ln 21 “The modelled difference increases if a coarser grid is used.”

Can the authors comment on the reasons for this? As discussed in the previous paragraph, a lower resolution should degrade the difference between Danum and Kunak

and thus result in a decreased difference.

We have added a substantial section to discuss the impact of resolution. We agree that this statement was confusing and it has been removed. New text includes: “Clearly, the finer the resolution selected for the output grid, the more trajectories must be run to ensure significance. So, with the NAME output grid used here, the Kunak grid box directly experiences coastal emissions while Danum does not. Higher resolution would likely degrade the statistics in figure 4 (fewer parcels per box) while a lower resolution would not allow us to distinguish between Danum and Kunak”.

p14979. Ln 4. The authors seem rather coy about discussing reasons for the modelled and measured diurnal variation. If this is found in the model (as well as the measurements) then clearly they have the means to explore this (photolysis/meteorology?), and this should be discussed since in their previous paper (O’Brien et al.) they allude to diurnal cycles being indicative of photochemical sources.

We are still investigating the diurnal variation. Either there should be a complete discussion or none; since our analysis is still ongoing, we think it is better at this stage to remove all mention of the diurnal variation and report this later, when the study is complete.

Ln 25. The model also captures a concentration gradient between the coast and inland, consistent qualitatively with the differences in bromoform measured at our two sites. I disagree: Figure 5 shows an insignificant difference between modelled bromoform at the two sites!

Discussion removed/changed. We now show pdfs of pTOMCAT results for inland Borneo and the coast. The figure shows that the observed behaviour is indeed reproduced qualitatively.

p14981. “p-TOMCAT is able to reproduce the magnitude of the bromoform measurements but only if the emission strengths used by Warwick et al. (2006) are reduced.”

The model can only reproduce the measurements at one site, and only if current assumptions about bromoform sources are substantially changed. Thus this statement is rather arbitrary. Later on, the authors state “The difference serves to emphasise the difficulty with using local measurements of short-lived halocarbons to attempt to infer global emissions.” I agree. In which case, I do not think it is appropriate to quote an estimated global source strength based on these measurements. Readers who only read the Abstract “the bromoform data are consistent with a lower global source (190 Gg Br yr⁻¹) than indicated by our recent measurements on Cape Verde (O’Brien et al., 2009)” may take this as a new CHBr₃ global estimate.

We fully accept these comments (and referee 2 makes similar points, also asking about the range of scenarios in Warwick et al (see below)). We now have additional model results where emissions are changed only in SEAsia. These too can reproduce the local observations but with, of course, a different global source again. We will discuss these in the context of the earlier Warwick study. An important part of the discussion will relate to understanding uncertainty.

The main conclusions that we have now reached can be summarized: The SE Asian emissions in Warwick, scenario 5, are too strong and overpredict surface bromoform in south East Asia So, we will recommend a reduction in these SE Asian emissions. However, reducing tropical coastal emissions everywhere by the same factor (which we did in the first draft of the paper, giving the global source of 190 Gg Br/yr)

underestimates free troposphere bromoform; the 190Gg number will now only be presented in our uncertainty discussion. No global number will be presented in the abstract. We have now also repeated the Warwick scenario 3 (just tropical ocean) and 5 (tropical ocean and tropical coastlines) calculations, modifying only the SE Asian emissions in line with our measurements. We find that with the limited observations available, it is not now possible to discriminate between these two and both scenarios have global emissions of around 400 GgBr/yr. However, the final important point that we now make is that while the local measurements in Borneo are a very important constraint on regional emissions, on their own they provide only a very weak constraint on global numbers.

Minor comments:

p14971 Ln 18: Law, Sturges et al., 2007). >Law and Sturges

OK

p14976 Ln 1-2 “We observed the bay at Kunak to be rich in macroalgae.” Please give some idea of the prevalent type of macroalgae.

OK – *Sargassum* sp

Anonymous Referee #2

The authors go on to compare their measurements with output from an Eulerian and a Lagrangian model, and conclude that both are unable to reproduce the high CHBr₃ enhancements at the coastal site, but that they need to scale down the emissions in the eulerian model by 2-6X to match the observed background levels.

The paper presents interesting data and the topic is appropriate for ACP. It is fairly well written. I see it as borderline in terms of whether the material and analysis as presented are sufficient to merit a standalone paper. The authors refer in the text to ongoing measurements, and that they now have 2 annual cycles of measurements from Borneo (the same inland site and a different coastal site). I find it confusing that the authors don't choose to include any of that information, and instead carry out a limited analysis based on just a few days of data.

The analysis of the longer data set is far from complete. We still think it better to try to write up this data from 2008 collected during the OP3 project.

Comments.

Abstract. “a lower global source of 190Gg Br/yr”. Given that the spatial and temporal distribution of bromoform emissions is not well-constrained, the authors should steer away from giving a global total based on these limited data in one location. Warwick et al. (2006) showed 7 emission scenarios with varying spatial distribution; picking one of these and scaling it down to arrive at a global flux estimate based on these few days of data is not really useful. The overall point the authors are making is fine “. . . point to the difficulty for short-lived species of extrapolating local measurements to a global scale”. Do any of the other Warwick scenarios perform better?

There is now a much more detailed discussion about the derivation of global emissions and associated difficulty/uncertainty (see response to ref 1 where this is covered in detail).

Section 2.2. The model description is inadequate, particularly for TOMCAT. Without saying what's driving the models, the data-model comparisons turn into a non-informative black box

exercise. You don't need to list all model details, but the salient ones should be included: how are the bromocarbon emissions computed / distributed? Resolution? Time step? How do we know the OH levels are reasonable?

We will add more discussion about the basic set-up of pTOMCAT

In several places there is a lack of numbers or statistics that are needed to back up statements in the text. For instance, - P14978, L20-21. "the concentrations at Danum are usually less than at Kunak, sometimes by a factor of two or more". Give numbers, e.g., X% of the time Danum is lower, and on average bromoform at Danum is Y% of Kunak. The statement is not clear from Figure 2. Visually they look very similar, and Danum is higher than Kunak on several occasions.

However it's not clear what you mean in the Figure 4 caption about the scaling. (Caption has been changed, see earlier).

P14978, L23-24, "slightly higher variability", what is that? Give SD or variance. Again this point is not apparent from the figure. - P14976, L27 "do not correlate with the bromoform peaks", this one is actually visible from the figure but you could still give an R value - P14980, L10, "produced identical measurements", need number, e.g. within 5%.

The referee wants a more convincing, quantitative demonstration concerning the difference and similarities in the data. We now show pdfs which we think convincingly show the data characteristics that we had previously described in words.

On a related note, P14979 L12-14, "The above has shown . . ." these actually haven't been shown convincingly, as per the previous comment.

Removed/rewritten.

P14980 L22-23, "substantial gradients between the coast and inland can be expected. . ." I can maybe see this in Fig 5, but not in Fig 4, need to be quantitative in both cases. Again, we have added pdfs from the models and changed the text.

P14981 L9 and L11, "good agreement", "agrees well", what does this mean?

Technical comments.

P14971, L23-24. Production is not controlled for all halocarbons, rephrase

OK

Section 2.1. Mention a few key details about the instrument, e.g. adsorbent(s) and column(s) used.

OK

P14977, L6, "and C2Cl4", phrasing is misleading since you don't look at C2Cl4 with the model

OK

P14978, L22, "the modeled difference increases if a coarser grid is used", but earlier you said "lower resolution would not allow us to distinguish between Danum and Kunak" so this is confusing, is it a typo?

See response to referee 1. What we wrote was clearly very confusing. We now have a substantial chunk of text discussing the impact of model resolution.