Review of Feng et al. 'Surface fluxes of bromoform and dibromomethane over the tropical western Pacific inferred from airborne in situ measurements'

General comments:

This manuscript describes an inverse modelling study designed to infer biogenic, oceanic emissions of two short-lived bromocarbons (bromoform and dibromomethane- CHBr3 and CH2Br2). The authors use an emission inversion setup consisting of a global chemistry transport model, a priori emission inventories for CHBr3 and CH2Br2, and aircraft measurements from two separate campaigns measuring both compounds over the tropical Western Pacific. The authors also carry out a short observing system simulation experiment to retrieve a set of known idealised emissions as a means of proving the efficacy of their inversion modelling setup. The authors conclude that the a priori emissions of CHBr3 and CH2Br2 are too high over this region, and find the a posteriori emissions of CHBr3 and CH2Br2 to be lower than the a priori emissions by 40% and 20%, respectively. They also conclude that assumptions in previous studies of a correlation in the emissions of CHBr3 and CH2Br2 cannot be supported based on the findings of this work.

The subject matter of the article sits well within the frame of ACP. In addition, the objectives of the scientific study and the design of the experiment (on the whole) mean that this work provides a useful scientific contribution on a topic (i.e. the biogenic, oceanic emissions of bromocarbons) where we have relatively poor understanding. It is welcome to see studies moving away from heuristic methods for inferring CHBr3 and CH2Br2 emissions towards using more robust methods. I therefore find that this paper is a welcome and much needed scientific contribution.

Overall, I find the article to be well written and organised. The scientific ideas are laid out in a clear and logical manner, and consequently one can follow the flow of ideas easily. The article also does quite well at justifying the methodological choices, although there is one major issue here that I will highlight below in the specific comments section. Unfortunately, this issue does have a direct bearing on my recommendation for publication. Separately, and as a more minor issue, I did find that the authors stopped short somewhat of some deeper discussion that I feel would help strengthen the article, and I will explain this in more detail below.

I therefore find that this has the potential to be a very good scientific article. However, I cannot recommend publication until the issues outlined in the specific comments section are addressed.

Specific comments:

1) The most significant problem I find in this study relates to the choice of Ordonez et al as the sole choice for priori emission inventory for both CH2Br2 and CHBr3 emissions.
    a. My first point relates to the CHBr3 a priori emissions. I fully recognise the challenges Ordonez et al. faced in creating an emission inventory using heuristic methods in a global model, and I fully respect the useful contribution Ordonez et al. have made to our understanding of VSLS emissions. However, we now have several studies (including this study and Ordonez et al. itself) that show that the Ordonez et al. CHBr3 emissions in particular are over estimated in the Western Pacific region.
        i. In fact Ordonez et al. (2012, ACP) itself in Figure 7 (the PEM-Tropics A, PEM-Tropics B, and TRACE-P panels) shows that their own model over estimates CHBr3 in the Western Pacific region when using their own emissions.
        ii. Ashfold et al. (2014, ACP) - another study employing a top-down method to infer VSLS emissions in the tropical Western Pacific - derived lower estimates of CHBr3 emissions in the tropics than Ordonez et al. Similarly, their retrieved

emissions (Fig. 6 panel D-F) show generally lower emission values than Ordonez et al., and the a posteriori emissions in this study (albeit in area not influenced by the emission inversion), in an overlapping region south of the Philippines.

iii. Hossaini et al. (2013, ACP) show the modelled tropical CHBr3 concentrations from the Ordonez et al emissions to be a consistently high outlier compared to those from the other emission data sets and observations (Fig.5 MLO, KUM, and SMO panels for the tropics; Fig. 7 30°N-30°S panels HIPPOS 1-5; and Fig. 11 all panels).

iv. Hossaini et al. (2016, ACP) Figs. 6 and 7 show that while using a much larger number of models, that the Ziska et al. emissions for CHBr3 generally outperform the Ordonez et al emissions in tropical locations.

v. In this study, the resulting simulated atmospheric concentrations of CHBr3 from the GEOS-CHEM CTM have a high bias as a result of using Ordonez et al. as the a priori.

vi. Unpublished work modelling seen by this reviewer that was presented at SHIVA meetings showing other models to overestimate CHBr3 concentrations in the western Pacific region when using the CHBr3 Ordonez et al emissions.

The conclusion I am making is not that the Ordonez et al emissions are too high in all regions of the world, but the preponderance of the evidence shows that they are too high over the important Western Tropical Pacific region considered in this study. I realise that the Ordonez emissions have been used recently by co-authors of this paper, and the link they have to ocean chlorophyll seems attractive, so perhaps they were a natural choice. However, based on the extensive evidence I presented above, I think that they were a poor choice (being the sole a priori tested). The a priori is an essential component of equation 2 necessary for a convergence to the global minimum of the cost function and for the best possible estimation of the emission state **c**. Therefore, selecting the best possible a priori emission dataset without large flaws is important for this study.

b. As a second point related to the first, I do not follow the logic of using the same published source for emission inventories of both CH2Br2 and CHBr3. One of the clear conclusions of the only comprehensive emission intercomparison study to date (Hossaini et al., 2013) was that no single emission inventory study was successful at producing good emissions for both compounds in question. The conclusions of Hossaini et al., were that Liang et al. provided the best estimates of CH2Br2 emissions over this region, and that Ziska et al. provided the best estimates of CHBr3 emissions. I do not think the authors should constrain themselves by picking the a priori emissions for two different compounds from the same published source when we know already that none of the published sources is able to satisfactorily give good results for both species.

c. I do have a further concern stemming from the fact that the a priori emission for CHBr3 seems to be too high and that the results of the OSSE show that the emission inversion setup has a tendency to overcompensate locally (to the observations). Simultaneously, the emission inversion seems to fail to significantly reduce errors in areas further away from the well observed areas of the domain. For the actual emission inversion, it seems entirely plausible that emissions could be being overly reduced in the well-observed area of the domain while remaining too high at the

western, northern and southern fringes. Can the authors please discuss how they think this issue affects their results.

Concluding my remarks on point 1), I strongly recommend, and as a condition of acceptance for publication, that in addition to running the emission inversion with Ordonez et al. as the a priori for both compounds, that the authors also run their emission inversion algorithm with Ziska et al. (CHBr3) and Liang et al. (CH2Br2) for the two compounds. Comparing this work to that of Ashfold et al. (2014, ACP), one can see that Ashfold et al. (2014, ACP) undertook a variety of emission inversion experiments (including changing their a priori) to test the setup of their system. These aspects of Ashfold et al. (2014, ACP) strengthened their work, and, similarly, this manuscript would also benefit from a similar effort.

2) Some key discussions seem to be missing including those of limitations of this study.
    a. It would have been nice to see a discussion of the prevailing meteorology during the period of study and an explanation linking this to the error reductions that we see in the OSSE results in Figure 3. Presumably, the error reductions are a function of the location of the observations and the origin of air masses arriving at the observation locations. An analysis similar to what I am suggesting was carried out in Ashfold et al. (2014) in their Figure 2, which allowed them to determine where there inversion setup was able to retrieve emission values. I realise this is perhaps easier in the Lagrangian framework of NAME, but the authors could draw upon the information in their meteorological inputs for GEOS-Chem to create a climatology of the winds and then make a discussion that would add useful context to the results and strengthen the paper.
    b. It would be good to see the authors try to connect the results of the OSSE, i.e., the spatially limited pattern of the error reduction, to the areas in the a posteriori CHBr3 emissions where we see the largest reductions in absolute emission values relative to the a priori. Given the evidence I present in point 1) above, I do not believe that the similarity in the spatial patterns in the OSSE error reduction and the area of reduced a posteriori emissions is coincidental. I think this implies that with greater spatial coverage in the aircraft observations that we would see reductions in the a posteriori emissions covering a larger spatial area. The authors should discuss this point, and also conclude that the spatial extent of the aircraft observations provides a limitation for this study.
    c. Further to point b., I do not find the a posteriori CHBr3 emission estimates outside of the region sampled by the observations (towards the N,S, W fringes of the domain) to be credible in light of the large reductions we see in the a posteriori compared to the a priori over the most sampled region. I am working on the assumption that the emissions are spatially correlated. Perhaps some discussion of this point in the context of the previous studies (e.g., those highlighted above in point 1) would help readers gauge the quality of the emission inversion in the areas on the N,S, W extremes of the domain where there is little information from the observations. This might also help readers understand the large gradients we see between the sampled and poorly sampled regions.
3) Figure colours in Figure 4 need greater differentiation. I struggled to differentiate the monochrome orange/brown tones. A set of panels representing the relative differences between the a priori and a posteriori emissions would also be of help.
4) I think it is necessary for the authors to include a discussion of the conclusions of Russo et al. (2015, ACP). Russo et al. (2015, ACP) made two conclusions relevant to the work in this study:

a. That it is difficult to infer emissions using aircraft measurements and coarse global models in the case where the emission distribution is heterogeneous in regions of strong convective activity.
b. That model resolution can affect the simulated distributions of CHBr3 in cases where the emissions distribution is heterogeneous.

The authors should include some discussion of these points and should explain how they present limitations for the current work, or why this points are not relevant to the conclusions in this manuscript.

5) It is important to note that the a posteriori emissions are more heterogeneous than the a priori. Therefore, following from Russo et al. (2015, ACP) and the discussions in point 4) above, the issue of model resolution could affect the simulated distribution of CHBr3 more significantly for the a posteriori emissions than for the a priori emissions. The authors have tested the impact of model resolution on the a priori emissions and found no effect. However, it seems plausible that model resolution could change the distributions of CHBr3 in the atmosphere more significantly for the a posteriori emissions given their greater heterogeneity. I recommend that the authors test this in a separate sensitivity study and present their conclusions.
6) It isn't clear to me that the mean bias between the mole fractions of observed and modelled CH2Br2 decrease from the a priori to the a posteriori simulations. The paper states this, but as it is written the bias changes from 0.01 +/- 0.14 to -0.1 +/-0.1. Can the authors please explain this result? Is this due to an overcompensation in the a posteriori emissions close the well observed region? According to the forward model section, a large fraction of the CH2Br2 originates from outside of the domain, and I imagine that in this case it is hard/impossible to infer those emissions with any reasonable specificity and overcompensation locally seems therefore to be a plausible explanation.

Technical comments:

Looking at Figure 4, it seems that the Ordonez et al and Ziska et al panels have been mislabelled in the caption whereby the Ziska emissions are described as being the Ordonez emissions and vice versa for the Ordonez emissions. Looking at Hossaini et al. (2013) ACP in figures 1 and 2 (and in fact the emission files themselves), I have checked the spatial patterns, and they seem to confirm this. Please can the authors check this themselves and confirm there is a mislabelling in the Fig. 4 caption? Please can the authors also check other instances of discussion of Ordonez and Ziska and verify that there a) there are no other mix-ups in the naming and b) that this is just a technical naming error.

Russo et al. (2015, ACP) is included as a reference but is not cited. Please check for other articles referenced but not cited.