Interactive comment on
"Oceanic bromine emissions weighted by their ozone depletion potential"
by S. Tegtmeier et al.

Anonymous Referee #1

Tegtmeier et al presents a quantitative estimate of the ozone depletion potential (ODP)-weighted emission calculation for the most abundant very-short-lived brominated compound, CHBr3. They concluded that (i) presently, the ODP-weighted CHBr3 emissions amount to ~50% of ODP-weighted anthropogenic emissions of CFC-11, and (ii) the ODP -weighted CHBr3 emissions will increase by 31% by 2100 due to increases in surface emissions and convective activity. While I have reserved opinions on the importance of calculation of ODP for CHBr3, which is predominantly of natural oceanic origin, I agree that its ODP information may be of use to some extent and the manuscript should be published after addressing the following comments.

We thank Referee 1 for his/her valuable comments. Please find below our response (in italic) to the comments as well as the according changes to the manuscript.

1. Section 2.1, 2nd paragraph. It would be good to add brief details of how the bottom-up emissions were derived in Ziska et al. In particular, it will be useful to show what are the major drivers of the 30% increase in VSLS emissions, whether it is due to increased surface temperature, winds, salinity, etc.

We have added the following text to section 2.1:
'For the time period 2006-2100, the global monthly mean emissions are calculated based on the monthly mean meteorological input parameters from CESM1-CAM5 and fixed atmospheric and oceanic concentrations from Ziska et al. (2013) following the parameterization of air-sea gas exchange coefficient from Nightingale et al. (2000). … These derived changes of the future VSLS emissions are only driven by projected changes in the meteorological and marine surface parameters, in particular, the by changes in surface wind and sea surface temperature. The respective contributions of wind and temperature changes to the future emission increase can vary strongly depending on the oceanic region (Ziska et al., in prep).'.

Further details of the predicted future emissions and the driving forces will be provided and discussed in the manuscript from Ziska et al. (Future emissions of halocarbons based on CMIP 5 model output fields) which will be submitted to ACPD within the next weeks.

2. Section 2.3, 2nd paragraph. I do not agree with the authors "active chlorine from CFC-11 will be impacted by changes in the stratospheric circulation in the same way as active bromine from CHBr3". The residual circulation will probably be sped up differently in different places. Since the short-lived and long-lived gases are released at different altitudes, the impact of CFC-11 and CHBr3 will be different, which will consequently affect the ODP calculation. However, I do agree with the authors that the impact of a speed-up circulation on CHBr3 ODP is small, compared to the other factors. Therefore, I suggest cutting the discussion short and ending with simply stating that the impact of the stratospheric residence time on CHBr3 ODP is expected to be small.

We agree with the referee that our discussion of the impact of changes in the Brewer-Dobson-Circulation on the VSLS driven ozone loss is not correct and have shortened the text as suggested above.

3. Ziska et al. 2013 emissions are found to be low-biased in the extratropics according to Hossaini et al. (2013). Although this bias will have a small, possibly negligible, impact on ODP-weighted CHBr3 emissions due to very small ODP in the extratropics, it still worth a brief discussion on the impact of this caveat on ODP-weighted emission calculation for CHBr3.

We have added a short discussion of this point. In particular, we have added the following text to Section 3:
'The evaluation of various $CHBr_3$ emission inventories from Hossaini et al. (2013) shows that in the tropics the best agreement between model and observations is achieved using the bottom-up emissions from Ziska et al. (2013). In the extratropics, however, the $CHBr_3$ emissions from Ziska are found to result in too low atmospheric model concentrations diverging from observations by 40 to 60%. … The distribution of the ODP-weighted emissions demonstrates clearly that $CHBr_3$ emissions from the NH and Southern Hemisphere (SH) extratropics have negligible impact on stratospheric ozone chemistry. Thus, the fact that the emissions from Ziska et al. (2013) might be too low in the extratropics (Hossaini et al., 2013) does not impact our results.'.

4. P14657, 2nd paragraph. Is it possible to find more literature information on how much of the CHBr3 emissions are currently due to aqua-farming? How much are they expected to grow (in percentage) in the coming decades? As stated by the authors, when it comes to ODP, it is indeed the anthropogenic component we care about.

We have added a discussion of the current and potential future $CHBr_3$ emissions from farmed seaweeds: 'In particular, aqua-farming used, among other things, for food production and CO_2 sequestering has started to increase as an anthropogenic VSLS source. Leedham et al. (2013) estimated tropical halocarbon production from macroalgae in the Malaysian costal region and suggest that only 2% of the local $CHBr_3$ emissions originate from farmed seaweeds. However, based on recent production growth rates, the Malaysian seaweed aquaculture has been predicted to experience a 6-11 fold increase over the next years (Phang et al., 2010). More importantly, other countries such as Indonesia, Philippines and China are known to produce considerably more farmed seaweed than Malaysia (e.g., Tang et al., 2011), but their contribution to the total anthropogenic VSLS emissions has not yet been assessed. The ODP of $CHBr_3$ demonstrates the high sensitivity of the South-East Asia region to growing emissions. Globally the highest ODP values (Figure 1b) are found in the same region where we expect future anthropogenic $CHBr_3$ emissions to increase substantially. An assessment of current and future seaweed farming activities including information on farmed species, fresh or dry weight macro algal biomass and incubation derived halocarbon production values is required to estimates the net oceanic aquaculture VSLS production.'

Minor comments:

The usage of emission vs. emissions is not very accurate and consistent throughout the manuscript. In many places, they are misused. Please carefully read through the manuscript and correct.

We have corrected the use of emission vs. emissions.

P14644, L22-24: -> a future climate. However, at the same time, it is reduced by less ...

We have changed the text.

P14645, L12: Should cite Carpenter & Reimann et al. (2014) (Chapter 1 of WMO 2014) instead of Chapter 1 of WMO 2011.

We have changed the citation.

P14646, L17: and not the -> but not the

We have changed the text.

P14646, L21-24: Change "Despite, ..." to "The ODP is traditionally ... However, some recent studies ... "

We have changed the text.

P14646, L26: Add "the" before long-lived halocarbons

We have changed the text.

P14647, L16: inside -> insight

We have changed the text.

P14647, L24-25: "While we focus our analysis on one VSLS and introduce the method and application exemplary for CHBr3", I understand what you mean here, but should consider rephrase

We have changed the sentence to 'The method and application are introduced for $CHBr_3$, within a case-study framework and can be applied to all VSLS where emissions and ODP are available at a spatial resolution necessary to describe their variability.'

P14648, L7: introduce -> introduced

We have changed the text.

P14649, L4: -> than the other CHBr3 ...

We have changed the text.

P14649, L8 & L17 & P14661, L17: Should this be Ziska et al. 2013? If it is Ziska 2015, it was not mentioned in the references.

This citation refers to a manuscript from Ziska et al., in preparation for submission to ACPD within the next weeks. We have changed the reference to Ziska et al., in prep.

P14650, L5: time scales play -> time scale plays

We have changed the text.

P14651, L14: delete "the" before tropospheric

We have changed the text.

P14652, L4: extent -> extend
We have changed the text.

P14652, L17: residence -> residence time
We have changed the text.

P14653, L22: -> the beginning and end
We have changed the text.

P14655, L17-21: Change "The potentially damaging effect of CHBr3" to "The impact of CHBr3". Are these the column integrated ODPs at the corresponding grid-cells?

Since we describe here not the actual but only the potential impact (only the impact CHBr3 would have if it would be really emitted from this location) we decided to change the text to 'the potential impact of $CHBr_3$ on …'. The ODP of the air parcels is calculated following their path through the troposphere and stratosphere (in a Lagrangian sense) and is in this Figure displayed at the location of the emission of the air parcel.

P14655, L21: delete "the" before "surface"
We have changed the text to 'the ocean surface'.

P14657, L2: "the mostly small ODP" – consider rephrase
We have changed the text to 'the overall relatively small ODPs'.

P14658, L16: extent -> extend
We have changed the text.

P14658, L18: -> we first analyze
We have changed the text.

P14659, L1: within these two months -> for June and December
We have changed the text.

P14664, L10: given -> due to
We have changed the text.

P14667, L25: CHBr3 from the surface -> transport of CHBr3 from the surface
The text should read 'CHBr3 delivery from the surface …'

P14667, L29: und -> and
We have changed the text.

P14668, L3: not well enough understood yet -> not understood well enough yet
We have changed the text.

P14668, L8: add "," after fields; "in order to derived" -> to derive
We have changed the text.

Interactive comment on
"Oceanic bromine emissions weighted by their ozone depletion potential"
by S. Tegtmeier et al.

Anonymous Referee #2

This paper presents ODPs for bromoform, and as such is interesting for the readership of ACP. This is obviously a complex topic but the authors have not adequately addressed or discussed this complexity. The science presented is incomplete. The introduction and abstract are poorly written, miss some significant points with regards to the complexity of convective changes with climate change and omit significant papers in the field concerning VSLS and climate. The contribution of bromoform due to anthropogenic sources (aquaculture) is not separated – and this would be possible, see below. Without these major issues in presentation and science in this paper being addressed I believe this work is currently not of sufficient quality for publication in ACP. The general approach and the aim of establishing emission weighted ODPs make this work very relevant and I encourage the authors to make these changes in order to improve the quality and scientific integrity of this work.

We thank Referee 2 for his/her valuable comments. We have rewritten the abstract and introduction, improved our discussion of future convective changes and included more information on anthropogenic sources. We believe that the paper has significantly improved providing now a more comprehensive study addressing all important aspects of the topic. Please find below our response (in italic) to the comments as well as the according changes to the manuscript.

[Leedham et al., 2013] state that 2% of current emissions are due to aquaculture in Malaysia, growing to 20% in the next decade. 94% of aquaculture in SEA occurs in Indonesia and Philippines – a more satisfactory discussion, and estimate for ODPs from aquaculture could be presented, making this paper's discussion of anthropogenic ODP's relevant for the Montreal protocol.

We have added a discussion of the current and potential future $CHBr_3$ emissions from farmed seaweeds to section 3: 'In particular, aqua-farming used, among other things, for food production and CO_2 sequestering has started to increase as an anthropogenic VSLS source. Leedham et al. (2013) estimated tropical halocarbon production from macroalgae in the Malaysian costal region and suggest that only 2% of the local $CHBr_3$ emissions originate from farmed seaweeds. However, based on recent production growth rates, the Malaysian seaweed aquaculture has been predicted to experience a 6-11 fold increase over the next years (Phang et al., 2010). More importantly, other countries such as Indonesia, Philippines and China are known to produce considerably more farmed seaweed than Malaysia (e.g., Tang et al., 2011), but their contribution to the total anthropogenic VSLS emissions has not yet been assessed. The ODP of $CHBr_3$ demonstrates the high sensitivity of the South-East Asia region to growing emissions. Globally the highest ODP values (Figure 1b) are found in the same region where we expect future anthropogenic $CHBr_3$ emissions to increase substantially. An assessment of current and future seaweed farming activities including information on farmed species, fresh or dry weight macro algal biomass and incubation derived halocarbon production values is required to estimates the net oceanic aquaculture VSLS production.'

We agree with the referee that an estimate of the anthropogenic fraction of ODP-weighted emissions would be a most interesting addition to the manuscript. However, a thorough assessment of the seaweed farming activities, which takes into account information on farmed species and farming location, is required before reliable estimates can be made. Such estimates will be subject of future studies.

Title – the paper does not deal with all oceanic bromine emissions' ODPs – so is overstated – change bromine to bromoform. This is particularly important for inorganic product gas washout, which is quite different for CH_2Br_2 relative to $CHBr_3$. [Liang et al., 2014] discuss how the different wet deposition processes of weaker convection favors CH_3Br versus CH_2Br_2 due to PGI washout. The results presented in this work are therefore only relevant for $CHBr_3$ and cannot be extended to all oceanic bromine as the authors have done.

We agree with the referee that the title was misleading and have changed it to 'Oceanic bromoform emissions weighted by their ozone depletion potential'.

Indeed, our results cannot be extended to all oceanic bromine; however, the method can be applied to all VSLS where emissions and ODP are available at a spatial resolution necessary to describe their variability. In the revised version of the manuscript we try to distinguish more clearly between the general concept introduced here (and the possibility of its application to all VSLS) and the specific results of our analysis that refer only to $CHBr_3$.

The abstract is far too general and requires tightening and quantification: in some places is very vague. For example the first sentence: "At present, anthropogenic halogens and oceanic emissions of Very Short-lived Substances (VSLS) are responsible for stratospheric ozone destruction." Is an oversimplification and ignores the roles of N2O, CO2, CH4, water vapour, aerosol etc in stratospheric ozone depletion, which are all very relevant in determination of future ozone (Chap 4, WMO ozone assessment, 2014). Line 6, page 14644: "Emissions of VSLS are, on the other hand, expected to increase in the future." and the next sentence can be combined and shortened. Or essentially lines 1- 8, page 14644 of the abstract could be removed as really introduction material and inadequately described in this abstract.

We agree with the referee and have rewritten the abstract as suggested.

Exemplary used throughout could be replaced within a case-study framework, as this is really what is presented in this paper.

The text has been changed as suggested.

Line 22, page 14644: larger convective activity is not descriptive enough or accurate (IPCC chapter 12 states less frequent, more intense convection with increased stability due to a higher tropopause). The explanation given by [Hossaini et al., 2012] is that while mid-tropospheric mass fluxes are depressed the higher tropopause in 2100 increases the 'depth' of the tropical convection and is accompanied by an increased mass flux to UT. This point needs to be discussed more completely in the paper – references within [Hossaini et al., 2012], [Rybka and Tost, 2014] and [Liang et al., 2014] would be helpful.

We agree with the referee that the term 'larger convective activity' is not sufficient. We have changed the term in the abstract to 'larger convective updraft mass flux in the upper troposphere'.

We have added the following text to section 7: 'More detailed evaluations demonstrate that the CESM1-CAM5 tropical convective upward mass flux is projected to decrease in the lower and middle troposphere (not shown here) in agreement with results from UKCA chemistry-climate model simulations (Hossaini et al., 2012). Contrary to the changes in the middle troposphere, the convective mass flux in the upper troposphere (above the 250 hPa level), is projected to increase in the future again in agreement with Hossaini et al. (2012). A higher extension of tropical deep convection has also been found in other model projections and global warming leading to an uplift of the tropopause has been suggested as the possible cause (Chou and Chen, 2010; Rybka and Tost, 2014). Overall, an increasing upward mass flux in the upper troposphere/lower stratosphere would lead to enhanced entrainment of $CHBr_3$ into the stratosphere, consistent with results from Hossaini et al. (2012) and Dessens et al. (2009), and thus to increasing ODP-weighted emissions.'

[Hossaini et al., 2015] is an extremely relevant reference for ozone depletion and the climate implications of VSLS that has been overlooked by the authors. This paper describes the vertical ozone loss seen by bromine, chlorine and iodine VSLS and how it is quite different than that of long-lived halogen species. The ozone losses in the Hossaini paper could be directly compared with the values found in this work. The [Hossaini et al., 2015] paper, combined with the [Tilmes et al., 2012] describe how the effect of VSLS on ozone is amplified under high stratospheric aerosol conditions. This is an important consideration for establishing ODPs for the VSLS that is neglected in the current work, especially as the background loading of stratospheric aerosol has increased in recent times [Solomon et al., 2011].

We agree with the referee that Hossaini et al. (2015) is an important reference that needs to be included in our paper (and was overlooked because it was published only three weeks before this study was submitted). We have added the text 'Brominated VSLS reduce ozone in the lower stratosphere with current estimates of a 3-11% contribution to ozone depletion (Hossaini et al., 2015) or a 2-10% contribution (Braesicke et al., 2013; Yang et al., 2014). Through the relatively large impact of VSLS on ozone in the lower stratosphere they have a radiative effect corresponding to a contribution of -0.02 W m-2 to global radiative forcing (Hossaini et al., 2015).' to the introduction. Since the ODP-weighted emissions used in our study is the overall effect of the bromine from $CHBr_3$ during its lifetime in the stratosphere and Hossaini et al. (2015) separate VSLS driven ozone loss in different stratospheric regions by combining all VSLS together, a direct comparison of the values is not possible.

With regard to the discussion of the impact of stratospheric aerosol we have added the text 'Finally, we do not consider potential future changes in stratospheric aerosol which could impact the contribution of VSLS to stratospheric ozone depletion (Salawitch et al., 2005; Sinnhuber et al., 2006). Variations in the background stratospheric aerosol loading (e.g., Vernier et al., 2011) are mostly attributed to minor volcanic eruptions (Neely et al., 2013). Since future volcanic eruptions are not accounted for in the simulations scenarios used here, we do not include the impact of natural aerosol variations. Suggested future geo-engineering would intentionally enhance the stratospheric aerosol loading and is predicted to increase the impact of VSLS on stratospheric ozone by as much as 2% at high latitudes (Tilmes et al., 2012). Such scenarios are not included in our simulations, but could effectively enhance the ODP of $CHBr_3$ due to an enhanced BrO/ClO ozone loss cycle in the lower stratosphere (Tilmes et al., 2012).' to chapter 8.

Page 14646, line 10 The simplistic model of [Schofield et al., 2011] shows that these uncertainties due to emissions inventories are inferior to those of modeled transport or wet deposition processes.

Schofield et al. (2011) prescribe boundary layer concentrations based on two representative examples but do not evaluate different existing emission inventories as it was done in Hossaini et al. (2013). We have added the references of Hossaini et al. (2013) and Schofield et al. (2011) to the sentence to give examples on where to find detailed information about the uncertainties.

Page 14647, lines 5-15 - ODPs is again introduced in this paragraph, this should come earlier and the repetition of the definitions removed (this would improve the flow of the introduction). Each paragraph should deal with a separate and new point.

While the earlier paragraph introduces ODP itself, this paragraph (lines 5-15) presents the concept of weighting surface emissions with the ODP. Since this concept has never been applied to short-lived substances before, we feel that it needs a separate paragraph. We have changed some of the wording to make the second paragraph more specific and the difference to the ODP paragraph before more clear.

Page 14651, line 15 (and elsewhere), only using the updraft mass-fluxes will lead to an error in the concentrations, as shown by [Frey et al., 2015] the downdraft mass fluxes are also very important 5-15% in determining composition at different levels. This will be important for oxidative capacities and ODPs as ozone is transported downwards from the stratosphere.

We absolutely agree with the referee that using the upward mass flux in order to derive the ODP is a simplified approach that does not take into account various processes such as changes in tropospheric chemistry, changes in the stratospheric residence time and, as pointed out by the referee, changes in the downward transport (e.g., from the UTLS into the troposphere). However, we aim to find a simple proxy that can be applied to climate model output taking into account the first order effects of future changes on the ODP. Based on the construction of the ODP proxy (simple linear fit relating the ODP at the emission location with the updraft mass flux at the same location) it is not possible to include the downdraft mass fluxes since they would not give an independent variable in the linear fit estimation. We have added 'While the downdraught mass fluxes can also impact (5-15%) the composition in the upper troposphere/lower stratosphere (Frey et al., 2015), they are not included in our proxy since there importance for the contribution of $CHBr_3$ *to stratospheric bromine is less clear and cannot be prescribed by a fit relation.' to section 4.*

Page 14652, line 24 The active chlorine is relevant for ozone loss predominantly in the polar regions, whereas active bromine is relevant at all latitudes (i.e. [Lee et al., 2002] – 80% of Bry is in radical form at midlatitudes). Therefore even if the assumption that stratospheric circulation would influence chlorine and bromine similarly holds (which it doesn't due to lower and upper branches of the Brewer-Dobson circulation being impacted differently by climate change), the ODPs of chlorine and bromine cannot be equated in this way – see also [Hossaini et al., 2015] for the altitude difference in ozone losses between VSLS and long-lived halogens.

We agree with the referee that our discussion of the impact of changes in the Brewer-Dobson-Circulation on the VSLS driven ozone loss is not correct and have shortened the discussion ending with simply stating that the impact of the stratospheric residence time on $CHBr_3$ ODP is expected to be small, as suggested by Referee 1.

Page 14647, line 16 – inside in should be insight into

We have changed the text.

Page 14648, line 7 introduce should be introduced

We have changed the text.

Discussion would benefit from a comparison with ozone assessment for CHBr3.

We have included a comparison from the 2014 ozone assessment in our discussion.