

**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 coastal 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 estimate 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 N_2O , CO_2 , CH_4 , 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 CHBr₃ 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 CHBr₃ 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 CHBr₃ 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 their 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 Br 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 VLSL 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 CHBr₃ 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.