

Dear Reviewers

We would like to express our sincere gratitude to the reviewers for your effort to improve our manuscript. Based on your comments, we've revised our manuscript accordingly with changed parts marked red.

The following is the point-to-point response with reviewers' comments in bold and the responses italic.

Comment on acp-2021-800

Paul J. Fraser (Referee)

Referee comment on "Potential environmental impact of bromoform from Asparagopsis farming in Australia" by Yue Jia et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-800-RC1>, 2022

Technical comments attached: Jia et al...

This is an important paper. CHBr₃ is a potent ODS and is produced in substantial quantities in the production of seaweed supplements to the diets of ruminants to suppress their CH₄ production. If adopted widely, this technology could substantially reduce ruminant CH₄ emissions which are a significant component of global CH₄ emissions. The paper address the important concept for short-lived ODSs that the impact on the ozone layer is dependent on the location of the emissions. The paper demonstrated the production of the necessary supplements to feed the global ruminant levels does not significantly deplete stratospheric ozone - the technology is 'ozone safe'.

I have a technical issue with the assumed/calculated levels of CHBr₃ resulting largely from coastal regions and natural seaweeds. I think the Zaffra et al. data, which are a compendium of CHBr₃ data from several laboratories, and are not intercalibrated (Zaffra et al. recognize this problem and have indicated it will be addressed in future studies) and potentially underestimate background levels of CHBr₃ in coastal regions. This seems to be the case in Tasmania (one of the study regions) where measured background CHBr₃ levels from the AGAGE program (not part of the Zaffra data, but arguable the best measured/calibrated CHBr₃ data set available) seem to be up to a factor of 3 higher than the Zaffra et al. data. Is this important? - the authors need to address this.

A: As shown in Eq (3) in the manuscript, the flux is calculated as the product of its transfer coefficient (k_w) and the concentration gradient (Δc), which is computed between the water concentration (c_w) and theoretical equilibrium water concentration (c_{atm}/H), the flux would be even weaker if c_{atm} increases, thus including such higher atmospheric background values in the Ziska methodology would not really increase the Ziska fluxes. On the other hand, if stronger coastal fluxes are applied, the conclusions will still hold.

To address this concern, we added several sentences in the discussion section "Another point to note is that Ziska emission potentially underestimate background levels of CHBr₃ in coastal regions (Ziska et al., 2013), e.g. CHBr₃ measurement in Cape Grim, which is close to Triabunna, shows much larger CHBr₃ mixing ratio (~1.5 ppt, Dunse et al., 2020). However, including such

higher atmospheric background values in the Ziska methodology would not really increase the fluxes as the flux is driven by air-sea gradient (Eq. 3). Our conclusions will still hold if stronger coastal emission is applied, as it will increase the background CHBr₃ mixing ratios.”

The authors need to review information on CHBr₃ atmospheric lifetime data and ozone impacts in the latest (2021) assessments of climate change (IPCC) and ozone depletion (UNEP)

A: The latest ozone assessment is 2018 version (the 2022 version isn't released yet), in which the CHBr₃ lifetime doesn't change too much compared to previous ones. We updated the information of CHBr₃ lifetime by adding the reference (Engel and Rigby et al., 2018).

Please also note the supplement to this comment:

<https://acp.copernicus.org/preprints/acp-2021-800/acp-2021-800-RC1-supplement.pdf>

Page 1, line 9: delete '4811' – no other co-author post-codes listed

A: deleted

Page 2, line 25: 'which contributes'

A: revised

Page 2, line 28: 'with its ozone depletion potential'

A: revised

Page 2, line 29: replace 'halogens' with 'halocarbons'

A: replaced

Page 2, line 35: 'significantly impact'

A: revised

Page 2, line 37: '0.02%' – significant figures implied by 0.016% probably not justified

A: revised

Page 2, line 37-38: 'The remains are relatively small' does not make sense

A: The sentence has been revised as "The impact of remaining farming scenarios is also relatively small".

Page 2, line 39: 0.5% (as above)

A: revised

Page 3, line 44: 'GHG emissions weighted by radiative forcing'

A: revised

Page 3, line 47: ‘emissions’

A: revised

Page 3, line 48-49: is this what you mean – ‘Total methane (CH₄) emissions from ruminant livestock contribute about 18% of total global CH₄ emissions’

A: Sorry for the confusion, the sentence is revised as “Total GHG emissions (e.g., CH₄) from ruminant livestock contribute about 18% of the total global carbon dioxide equivalent (CO₂-eq) inventory...”

Page 3, line 51: quote GWP and lifetime data for CH₄ from more recent IPCC assessments: IPCC Climate Change 2021

A: New reference of the sixth IPCC assessment is quoted.

Page 3, line 56: ‘inefficient digestion process’

A: revised

Page 4, line 74: Carpenter and Liss, 2000

Page 4, line 77-78: Engel, Rigby et al., 2018

Page 4, line 81: Carpenter, Reimann et al., 2014

A: All the reference issues above are revised.

Page 4, line 83-84: in the lower and middle stratosphere

A: revised

Page 4, line 85: Black et al. 2021 not in reference list

A: added

Page 5, line 115: For an effective

A: revised

Page 5, line 115: 0.4%? Can two significant Figures (0.38%) be justified; could the experiments distinguish between results for 0.38% and 0.40?

Page 5, line 119: 3.5 x 10⁴ Mg – 5 significant figures??

Page 5, line 121: 2.3 x 10⁴ Mg – 5 significant figures??

Page 6, line 135: 1.2 x 10⁴ Mg

Page 6, line 139: 3.5 x 10⁴ Mg

A: The significant figures all through the manuscript were revised. Rounded values with at most two significant figures are used for description parts, except for specific calculations (e.g. Table 1).

Page 6, line 147: Yong et al. (2013)

A: revised

Page 7, line 166: which farms are actual and theoretical?

A: the caption of figure 1 is revised as “Locations of actual Asparagopsis farms in Geraldton, Triabunna, Yamba, and theoretical farms in Darwin.”

Page 8, line 171: 1.2×10^4 Mg

A: revised, see above

Page 9, line 180: listed as Magnusson et al. 2020 in reference list

A: corrected.

Page 9, line 185: 1650 ng, the Paul et al. release is quoted to 2 significant figures: 1100 ng

Page 9, line 187: OK, 1600 ng, ignore above

Page 9, line 191: were used in this study

A: revised

Page 10, line 211: Mass et al references are listed as 2019 and 2021

A: corrected

Page 10, line 228: inter-? Interpolation?

A: revised as “...filled by interpolating and extrapolating...”

Page 11, line 247: 3.5×10^4 Mg

A: revised

Page 12, line 285: Montzka, Reimann et al. 2010

A: revised

Page 14, line 298: to that of the reference substance CFC-11 (CCl₃F) on a mass emitted basis

Page 14, line 300: long-lived halocarbons

Page 14, line 304: from the same unit mass emission of CFC-11

A: revised

Page 14, line 307: CHBr₃

A: revised

Page 15 line 326: the ODP values applied here

A: revised

Page 15, line 328: Engel, Rigby et al.

A: revised

Page 15, line 329-330: impact on the comparison.....ODP weighted emissions presented here.

A: revised

Page 18, line 324: to simulate the enhanced atmospheric CHBr₃ mixing ratios (above natural background) for each...

A: revised

Page 18, line 395: Background CHBr₃ levels are calculated.... These background levels derived from Ziska et al. need to be discussed. Do the authors use the latitudinally averaged data (Figure 2 of Ziska et al.). It would be instructive to compare the background CHBr₃ levels from Ziska et al. assumed for Triabunna, Tasmania (about 0.5 ppt?) to publicly available (and published) observations at Cape Grim, Tasmania (annual average about 1-2 ppt). The Ziska data compendium are from various laboratories but are not intercalibrated. The Triabunna background CHBr₃ level could be as high as 2 ppt – what difference would this make to the flux calculations?

A: To address this comment, we added a short discussion in Section 5, see the response to the main comment above.

Page 18, line 398: Figure S1 compares calculated CHBr₃ mixing ratios due to Asparagopsis farming at GTY and Darwin compared to appropriate Ziska mixing ratios (need to state latitude of Ziska data). The Figure shows mixing ratios only, not emissions, so need to state this and then say what this implies about emissions. The background surface CHBr₃ mixing ratios in Figure S1 are 0.01 ppt, this an order of magnitude lower than the Ziska data for Darwin latitudes (minimum 0.5 ppt). Am I missing something here?

Page 18, line 400: Compared to Figure S1, Figure 4 has Ziska coastal surface mixing ratios of 0.05 ppt, 5 times Figure S1?

Page 18, line 409: Figure 5 shows Ziska coastal CHBr₃ mixing ratios of 0.1 ppt or higher, at least 2 times Figure 4 and 10Xx Figure S1??

A: The differences mentioned in the above 3 comments are from the way how the results are shown (mainly because of averaging). Figure 5 shows the spatial distribution of CHBr₃ mixing ratios, while Figure 4 and Figure 1S show regional averaged mixing ratios, resulting in smaller mean values. The mixing ratios due to Ziska emission in Figure 4 and Figure 1S (bottom panels) are

actually the same, we chose different color scale in the two figures to make the signals of other scenarios more “visible” as the emission magnitudes in Figure 4 are 30 times as in Figure 1S.

Page 21, Figure 4 caption: Altitude-time cross-sections of CHBr₃ mixing ratio averaged over [...] from a)...

A: revised

Page 22, line 456: I would have thought that the ODP distribution also depends on the surface location of the CHBr₃ emissions (as well as seasonal transport, location of ITCZ etc). Which emission scenario (strength, location) is used to calculate the ODP distribution?

A: The ODP of VSLS is a function of time and location of the emissions, the seasonal transport and location of ITCZ have already implied the location-dependence. The ODP values used in our study are taken from Pisso et al. (2010), in which a map of ODP was created by calculating the ODP for each emission grid globally. The strength of the emission does not matter in calculating the ODP for VSLS as it is the fraction of parcels into the stratosphere released from a certain grid.

Page 23, line 479-480: 1.1%, 2.9%

A: revised

Page 24, line 500: ...ODP of CHBr₃ in January.... Specify emission scenario?

A: In Figure 7, the ODP values are from Pisso et al. (2010), the caption has been revised as “Figure 7 Spatial distribution of the ODP in January and July from Pisso et al. (2010), plotted with interval of 0.01”. The corresponding description in section 4 is also revised by adding the reference.

Page 25, line 514: 3.47

A: revised

Page 25, line 520: CHBr₃

Page 26, line 527, 532, 542: CHBr₃

A: corrected

Page 26, line 550: laboratory scale

A: revised

Page 28, line 608: Battaglia not cited (I may have missed it)

Page 28, line 614: Black et al. 2021 not listed

Page 29, line 663: Herrero et al 2016 not cited (I may have missed it)

Page 31, line 728: Machado et al. 2014 not cited (I may have missed it)

Page 31, line 746: Marshall et al. should be listed before Mata et al.

Page 32, line 775: Montzka, Reimann et al.

Page 33, line 840: Wuebbles et al. 1983 not cited (I may have missed it)

A: These reference issues have been addressed with missing references added, not cited reference removed, format and order adjusted.