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.

Review of the manuscript "Potential environmental impact of bromoform from Aspargopsis farming in Australia" by Jia et al., ACPD, 2021.
Rafael Pedro Fernandez (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-RC2, 2022
The paper presents modeling experiment to evaluate how one of the proposed feeding management alternatives to reduce CH4 emissions from ruminant livestock (i.e., Aspargopsis farming) could impact on the stratospheric ozone layer due to the by-product formation of bromoform (CHBr3). This species is a very short-lived species (VSLS) with a mean lifetime of 17 days in the atmosphere, and consequently, the CHBr3 impact on stratospheric ozone depends on the superposition of source strength and location with the efficiency of convective transport. The paper proposes a multiple set of realistic local and global scenarios, as well as the occurrence of some improbable extreme episodes affecting the Australian coast, to evaluate a representative range of the overall ozone depletion potential (ODP) of bromoform emissions from oceanic and terrestrial cultivation approaches, and compare them with the impact of coastal natural bromoform emissions.
The work is very well-planed and provides a realistic and clear evaluation of the magnitude of one of the environmental consequences of promoting Aspargopsis production in Australia, and determine that even in the worse possible scenario, the negative impact of the additional farming-released bromoform are very small in comparison with the natural contribution from the ocean. The methodology and results are generally well presented, although some clarification is required as described below. I suggest the paper is accepted for publication after the issues/comments in the attached file have been solved.
Please also note the supplement to this comment:
https://acp.copernicus.org/preprints/acp-2021-800/acp-2021-800-RC2-supplement.pdf

Main Comments:
1a. Ozone Depleting Potentials (ODPs): Concept and Implications
Section 2.5 briefly describes the ODP concept and how it has been adapted to evaluate the ODP impact of VSLS due to their variable distribution in the troposphere. However, given the importance of the ODP fields used to determine the bromoform ODP-weighted emissions presented in this work, I found that more details (and results discussion in Section 4) should be given. In particular, the authors based their analysis on the ODP spatiotemporal study

performed by (Pisso et al., 2010) using the same FLEXPART model, but no mention is provided about other approaches to determine the Stratospheric ODP (SODP) for long-lived species that are known to affect both tropospheric and stratospheric ozone (Claxton et al., 2019; Zhang et al., 2020), and why it is important to distinguish the tropospheric and stratospheric ozone impacts of $CHBr_3$.

A: The ODP values calculated in Pisso et al. (2010) are SODP as only the impact on stratospheric ozone was considered. To address this comment, we revised the corresponding part in the introduction by adding "Once released into the atmosphere, degraded halogenated VSLSs can catalytically destroy ozone in the troposphere and stratosphere, thus drawing them considerable interest (Engel and Rigby et al., 2018; Zhang et al., 2020).".

Also, in Sec 2.5, the following sentences are added "The ODP for VSLSs can be derived from chemistry-climate or chemistry transport models simulating the changes of ozone due to certain compound (Claxton et al., 2019; Zhang et al., 2020). The trajectory-derived ODP of VSLSs such as $CHBr_3$ is calculated as a function of location and time of the potential emissions (Brioude et al., 2010; Pisso et al., 2010). As for the traditional ODP concept, the time and space dependent ODP describes only the potential of a compound but not its actual damaging effect to the ozone layer and is independent of the total emissions. It is noteworthy that many VSLSs including $CHBr_3$ can impact ozone in the troposphere and stratosphere. As ODPs are used to assess stratospheric ozone depletion only, the contribution of VSLSs to tropospheric ozone destruction needs to be excluded when calculating their ODP (Pisso et al., 2010; Zhang et al., 2020). The trajectory based ODP from Pisso et al. (2010) used in this study, considers only the impact of $CHBr_3$ on the stratospheric ozone instead of the ozone column."

Page 15, Lines 326-329 is the only place in the text where I found explicit mention that the product gas contribution of VSLS degradation is not being considered, which is reasonable as the proposed methodology considers only the exponential decay of the emitted source gases. However, this should be at least highlighted again in the conclusions and if possible, an estimation of the magnitude of the neglected tropospheric impact of VSLS product gases and/or how the modeling ozone changes depend on the treatment of VSLS product gases (i.e., Fernandez et al., 2021) could be given.

A: In Section 5, we added the following discussion to highlight the missing of product gas contribution "The ODP used in this study, does not include the impact of VSLS product gases. Previous modelling studies have highlighted the role of product gas treatment and their impact on the stratospheric halogen budget (e.g., Fernandez et al., 2021). Including product gas entrainment can lead to up to 30% larger ODP values for $CHBr_3$ (Engel and Rigby et al., 2018; Tegtmeier et al., 2020), thus the ODP-weighted emissions presented here can be up to 30% larger. However, this does not affect our assessment of the potential importance of cultivation induced $CHBr_3$ as the ratios of the impact of each scenario compared with the global ODP-weighted emission remain the same."

1b. Ozone Depleting Potentials (ODPs): Methodology

The ODP for bromoform is computed by comparing the ozone destruction of CHBr3 compared with the ozone destruction produced by an equivalent mass of CFC-11. However, no CFC-11 sensitivity is mentioned to have been performed for this study. Thus, it is not clear if Fig. 7 is a direct result of the modeling simulations performed in this work, or it is taken from Pisso et al., (2010). If the later is the case (which I believe it is), then, this should

be expressed more clearly in the text and proper reference to this study should be given in the caption of Fig. 7.
A: In this study, the ODP is taken from Pisso et al., (2010). To clarify this, caption of Fig 7 is revised as "Spatial distribution of the ODP in January and July from Pisso et al. (2010), plotted with interval of 0.01". And the beginning of Sec 4 is also revised as "The ODP distribution from Pisso et al. (2010) for the region around Australia… shown in Figure 7".

Page 14, Lines 311-313 explicit says that "ODPs for VSLSs are calculated by means of combining two sources of information: one corresponding to the slow stratospheric branch and the other to the fast tropospheric branch of transport". First, how the tropopause location is determined in the study? Second, is it possible to quantify the contribution of these two branches, and could this be taken as an approximation of the tropospheric and stratospheric influence of CHBr3 farming emissions? Note that one of the main results of the paper is that ODP-weigthed CHBr3 contribution from Aspargopsis farming would be, at most, less than 1% of the natural CHBr3 value (i.e., the Ziska_Coast scenario); thus properly showing how the ODP values were computed for this particular VSLS should be clear.
A: In Pisso et al. (2010), the tropopause is the WMO thermal tropopause was applied. The contributions of the two branches could be quantified but cannot be used as the approximation of the impact on troposphere and stratosphere. While the transport pathways in the troposphere and stratosphere are included, the ODPs are only calculated for the stratosphere chemistry (see also the response to comment 1a).

Minor Comments:
GENERAL: The number of significant digits used when reporting numbers should be revised throughout the hole text.
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).

P2,L36: What do you mean by "the remains are relatively small"?
A: Sorry for the confusion, the sentence has been revised as "The impact of remaining farming scenarios is also relatively small".

P2,L37: "less than 0.016%" … is this significant different to less than 0.02%?
A: The value is revised as 0.02%, also see the response to the number of significances.

P2,L39: "by 0.48%" … of its initial value, or up to 0.48%?
A: revised as "up to ~0.5%"

P4,L88: "In consequence, the environmental impact of CHBr3 … needs to be explored and elucidated". As detailed in the main comment, the authors should explicit mention that VSLS influence both the troposphere and stratosphere, and that here only the stratospheric impact is considered.
A: The corresponding part in the introduction is revised as "Once released into the atmosphere, degraded halogenated VSLSs could catalytically destroy ozone both in the troposphere and

stratosphere, thus drawing them… explored and elucidated. In this study, only the impact on the stratosphere is considered."

P4,L94: I found the paper very informative not only to industry, but also to policy makers and the scientific community.
A: Thanks for the reviewer's recognition. The sentence is revised as "Specific objectives were to inform the industry, policy makers, as well as the scientific community on:…"

P5,L119: 3.4674 x 10^4. Does this number have 5 significant digits? Please clarify and make it consistent throughout the text.
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).*

P5,L128-130: How did you get the 30 times scaling factor to extrapolate from Australian Aspargopsis production to Global production? And how did you get the 1 Tg DW value? (I could not get that value by multiplying the informed data … I must have missed something).
*A: As described in the assumption, the Australian feedlot and dairy industries that adopted Aspargopsis is approximately equivalent to 1% of the global feedlot and dairy herds (assumption iii), and 30% of the global feed base would adopt Aspargopsis (assumption i), which results in a scaling factor of 30. As the current annual yield is 34674 Mg DW, the scaled yield to the global scale will be 34674*30 = 1040220 Mg (~1 Tg).*

Figure 1: The lat,lon region shown in the Figure is smaller than the rectangle used for computing the average of CHBr3 mixing ratio in Figs. 4 and 6.
A: The larger rectangle in Figure 1 is for the convenience of better showing the locations of the farms on the map, otherwise, the locations would be almost at the edge of the plot.

P10,L233: Considering extending the subsection title so it includes the description of the different scenarios. In addition, by looking at Table 1 it is evident that the study was performed for meteorological conditions of year 2018 … But I could not find where in the text this is described (I might have missed it).
A: The L246 is revised as "We conduct FLEXPART simulations for year 2018 with different emission scenarios as…" to highlight that the study performed for 2018. The subsection title is changed to "2.4 Emission Scenarios for FLEXPART Simulations".

P11,L263 and Table 1: The total CHBr3 emission within the background scenario considers the well-established Ziska emission inventory, and is mentioned to consider "all 1 x1 grid cells directly neighboring the coastline", which accounts for 3109 Mg (Table 1). How large are the Ziska emissions for a small region of the size of area of Geraldton, Triabunna or Yamba? Similarly, how large are the Ziska_coast emissions if they are compared to the total Ziska emission on the Australian domain [10 -45 S, 105 -165 E] if both coastal and open-ocean grid-cells are considered?
A: The Ziska emissions on the domain for coastal and open ocean (also with shelf) are 3109 Mg and 2047 Mg, respectively. It is not reasonable to compute the Ziska emission on the locations of farming as some farms are terrestrial. However, if we assume all the farms are Geraldton-like (i.e.

all grown in the open ocean), the Ziska emission in Geraldton, Yamba and Triabunna will be 843 Mg, 295 Mg, and 676 Mg, respectively. To address this comment, these numbers due to Ziska emission are added to Section 2.4 with the sentence "Note that it is not reasonable to compute the Ziska emission on the locations of farming as some farms are terrestrial. However, if we assume all the farms are Geraldton-like (i.e., all grown in the open ocean), the Ziska emission in Geraldton, Yamba and Triabunna will be 843 Mg, 295 Mg, and 676 Mg, respectively."

P14,L311: The 20 days lifetime of the VSLS species considered in Pisso et al., (2010), should be mentioned here.
A: The sentence is revised as "…VSLS with a lifetime of 20 days, which is very similar to that…"

P15,L321-323: "In this study, we present the ODP-weighted emissions, which combine the information of the ODP and surface emissions and are calculated by multiplying the CHBr3 emissions with the trajectory-derived ODP at each grid point". Does Pisso et al., (2010) provide independent ODP values for each model grid-point and individual trajectory? Please see my main comment regarding this point.
A: The ODP values from Pisso et al. (2010) are calculated for each grid on the emission map. The values for each grid are given instead for every trajectory.

P15,L344 and Fig. 3a: The figure is fine, and is clear that the annual emission for the different growth periods are equivalent, but the text seems to imply that this is a new result of the study. However, these equivalent values is just a confirmation of the assumed condition that all farming scenarios for Australia must have the same total emission. This should be clarified in the text.
A: To clarify this, we replaced "reveal" with "show", and "indicating" is replaced with "confirming" to reinforce that this part is a conformation.

P16,L363: "which leads to emissions of 27 Mg (0.1 Mmol) CHBr3 per year for the targeted final yield". How do you relate this 27 Mg CHBr3 per year with the aprox. 9 Mg CHBr3 annual emission derived from Fig. 3a? Shouldn't this values be identical? Is it needed to multiply by the bromine atomicity of bromoform (3)?. Please make it clear.
*A: The emission 9Mg from Fig 3a is based on 11558 Mg DW, which is the total annual yield of one farm. For three farm locations: Geraldton, Yamba, and Triabunna, the total annual emission will be 9Mg *3 =27 Mg.*

Figure 3 caption: "… under different growth rates and similar initialbiomass and growth period". Please make the caption as informative as possible.
A: The caption is revised as "Figure 3. The annual release of $CHBr_3$ (Mg yr^{-1}) from: a) same growth rate (5%) for different growth periods; and b) under different growth rates but with same initial biomass, both a) and b) are obtained with a total annual yield of 11558 Mg DW."

P19,L430: "and signals with comparable magnitudes are found at 15 km". The magnitudes are comparable, but I expect this signals affect much smaller regions due to the localized source. Is this the case? If so, please make it explicit for the reader.
A: The sentence is revised as "...and signals with comparable magnitudes, though with smaller coverage, are found at 15 km."

Figs. 4 and 5: Is the color scale maximum value correct? i.e. 0.05 ppt for Fig. 4 and 0.10 ppt for Fig. 5? How large are the maximum values within the MBL? I would expect them to be much larger than the maximum value of the scale.
A: The difference of the color scales between Fig 4 and 5 is due to the averaging over the domain, which leads to smaller mean values. Also, for the convenience of comparing the signals due to each emission scenario, especially in the free troposphere and stratosphere, we chose smaller color scale in Fig 4. The maximum value due to Ziska_Coast in the MBL is ~0.15 ppt.

The caption of Fig. 4 should also explicitly indicate that it refers to Global scenarios.
A: The caption for Figure 4 is revised as **"***Altitude-time cross-sections of $CHBr_3$ mixing ratio averaged over [10°-45° S, 105°-165° E] from Global Scenarios: a) GTY_O60_30x, b) GTY_O96_Jan_30x, c) GTY_O96_Jul_30x, d) Darwin_O60 _30x, and Background Scenario: e) Ziska_Coast."*

Figure 7: If the units of the scale is a relative value between 0 and 1, please make it explicit.
A: The caption of Figure 7 is revised as "Spatial distribution of the ODP in January and July from Pisso et al. (2010), plotted with interval of 0.01"

Figure 8: The bottom-most bar presenting values for the Global Emission, for which of the global scenarios apply?
A: The Global Emission is taken from Tegtmeier et al. (2015) as a reference number, not from the emission scenarios in this study. To avoid the confusion, the caption of Fig 8 is revised as "Figure 8. The ODP-weighted emissions of $CHBr_3$ for Global Scenarios (GTY_O60_30x and Darwin_O60_30x), Australian Scenarios (GTY_O60 and Darwin_O60), Coastal Australian emission (Ziska_Coast), and global ODP-weighted emission for 2005 taken from Tegtmeier et al. (2015) as a reference, note that the x-axis is exponential."

Language editing comments:
GENERAL: A language style revision should be performed to the whole text (as well as figure captions), mainly on the unification of past, present and future terms (is, was, will) into a common verbal tense.
A: We've revised the verbal tense all through the manuscript.

P2,L26-30: Split the sentence.
A: The sentence is split as "In this study, we focus on the impact of $CHBr_3$ on the stratospheric ozone layer resulting from potential emissions from proposed Asparagopsis cultivation in Australia. The impact is assessed by weighting the emissions of $CHBr_3$ with its ozone depletion potential (ODP), which is traditionally defined for long-lived halocarbons but has been also applied to very short-lived substances (VSLSs)."

P2,L30: DW acronym is not used again in the abstract.
A: The acronym DW is removed.

P3,L48: Two blank spaces.
A: revised

P3,L64: rephrase "showed the most potential for CH4 production decrease".
A: The sentence is revised as "…showed the most potential for reducing CH_4 *production…"*

P9,L189: What do you mean by "as the farming aims at high yielding CHBr3 varieties"?
A: The algae in our study are varieties with higher CHBr3 content and yield than those in the wild. To avoid confusion, the sentence is revised as "These content and release rates are higher than those for wild stock algae (Leedham et al., 2013; Nightingale et al., 1995) as the farming aims at algae varieties with high $CHBr_3$ *yield."*

P10,L214: "the gradient is between" … it is computed between? It is computed considering …?
A: revised as "The gradient is computed between…"

P15, L323-324: "The ODP-weighted emissions provide insight in where and when CHBr3 is emitted that impacts stratospheric ozone (Tegtmeier et al., 2015)". Not sure if the sentence is properly written. Please rephrase.
A: The sentence is rephrased as "The ODP-weighted emissions provide insight into key factors of $CHBr_3$ *emission (i.e. where and when* $CHBr_3$ *is emitted) that impact stratospheric ozone (Tegtmeier et al., 2015)."*

P15,L329-330: "but has no large impact on the here presented comparison of global ODP-weighted CHBr3 emissions with farm-based ODP-weighted CHBr3 emissions.". Please rephrase.
A: The sentence is rephrased as "but has no large impact on the comparison between global ODP-weighted $CHBr_3$ *emissions and farm-based ODP-weighted* $CHBr_3$ *emissions presented here."*

P19,L423: The authors use the terms "destroy" to refer to the impact of cyclone Joyce on the Australian coast. Please consider using a different wording (here and elsewhere).
A: The term "destroy" is replaced by "farm could be totally damaged by cyclone Joyce"

P23,L488: remove "again"
A: removed

P25,L517,520: (here and elsewhere). Use subindex for 3 in $CHBr_3$**.**
A: revised

P26,L525: "The local CHBr3 emissions from the Asparagopsis farms could be larger than emissions from coastal Australia." The term "local" here is correct, but seems hidden in the sentence and could be reinforced.
A: The sentence is revised as "The $CHBr_3$ *emissions from the localized Asparagopsis farms".*

References:

Claxton, T., R. Hossaini, O. Wild, M.P. Chipperfield, and C. Wilson, On the regional and seasonal ozone depletion potential of chlorinated very shortlived substances, Geophys. Res. Lett., 46(10), 5489–5498. doi:10.1029/2018GL081455, 2019.

Fernandez, R.P., J.A. Barrera, A.I. L pez-Nore a, D.E. Kinnison, J. Nicely, R.J. Salawitch, P.A. Wales, B.M. Toselli, S. Tilmes, J.-F. Lamarque, C.A. Cuevas, and A. Saiz-Lopez, Intercomparison between surrogate, explicit and full treatments of VSL bromine chemistry within the CAM-Chem chemistry-climate model, Geophys. Res. Lett., 48(4), doi:10.1029/2020GL091125, 2021.

Pisso, I., P.H. Haynes and K. S. Law, Emission location dependent ozone depletion potentials for very short-lived halogenated species, Atmos. Chem. Phys., 10, 12025–12036, https://doi.org/10.5194/acp-10-12025-2010, 2010.

Zhang, J., D.J. Wuebbles, D.E. Kinnison, and A. Saiz-Lopez, Revising the ozone depletion potentials metric for short-lived chemicals such as CF3I and CH3I. J. Geophys. Res. Atmos., 125, e2020JD032414. https://doi.org/10.1029/2020JD032414, 2020.