Potential environmental impact of bromoform from Asparagopsis farming in Australia

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Abstract

To mitigate the rumen enteric methane (CH$_4$) produced by ruminant livestock, *Asparagopsis taxiformis* is proposed as an additive to ruminant feed. During the cultivation of *Asparagopsis taxiformis* in the sea or in terrestrial based systems, this macroalgae, like most seaweeds and phytoplankton, produces a large amount of bromoform (CHBr$_3$), which may contribute to ozone depletion once released into the atmosphere. In this study, the impact of CHBr$_3$ on the stratospheric ozone layer resulting from potential emissions from proposed *Asparagopsis* cultivation in Australia is assessed by weighting the emissions of CHBr$_3$ with the ozone depletion potential (ODP), which is traditionally defined for long-lived halogens but has been also applied to very short lived substances (VSLSs). An annual yield of $\approx 3.5 \times 10^4$ Mg dry weight (DW) is required to meet the needs of 50\% of the beef feedlot and dairy cattle in Australia. Our study shows that the intensity and impact of CHBr$_3$ emissions varies dependent on location and cultivation scenarios. Of the proposed locations, tropical farms near the Darwin region are associated with largest CHBr$_3$ ODP values. However, farming of *Asparagopsis* using either ocean or terrestrial cultivation systems at any of the proposed locations does not have potential to impact the ozone layer. Even if all *Asparagopsis* farming was performed in Darwin, the emitted CHBr$_3$ would amount to less than 0.016\% of the global ODP-weighted emissions. The remains are relatively small even if the intended annual yield in Darwin is scaled by a factor 30 to meet the global requirements, which will increase the global ODP-weighted emissions by 0.48\%
1. Introduction

Livestock is responsible for about 15% total anthropogenic greenhouse gas (GHG) emissions (Gerber et al., 2013), ranking it amongst the main contributors to climate change. The global demand for red meat and dairy is expected to increase >50% by 2050 compared to 2010 level, thus mitigation measures to reduce the GHG emission from the global livestock industry are in high demand (Beauchemin et al., 2020). Total GHG emissions from ruminant livestock contribute about 18% of the total global carbon dioxide equivalent (CO$_2$-eq) inventory as CH$_4$ (Herrero and Thornton, 2013). With a global warming potential 28 times higher than carbon dioxide (CO$_2$) and a much shorter lifetime (~10 years, IPCC, 2014), ruminant enteric CH$_4$ is an attractive and feasible target for global warming mitigation.

Enteric CH$_4$ from ruminant livestock is produced and released into the atmosphere through rumen microbial methanogenesis (Morgavi et al., 2010). Methanogenic archaeea (methanogens) intercept substrate CO$_2$ and H$_2$ liberated during bacterial fermentation of feed materials (Kamra, 2005), and during this inefficiency of the digestion process (Herrero and Thornton, 2013; Patra, 2012), methanogen metabolism leads to reductive CH$_4$ production and loss of feed energy as CH$_4$ emissions. To abate enteric methanogenesis, different strategies such as feeding management and antimethanogenic feed ingredients, have been proposed and assessed (e.g., Moate et al., 2016; Mayberry et al., 2019; Beauchemin et al., 2020). Some types of macroalgae have been demonstrated to mitigate production of CH$_4$ during in vitro and in vivo rumen fermentation significantly (Kinley and Fredeen 2015; Li et al., 2018; Kinley et al., 2020; Abbott et al., 2020). Among the different macroalgae species, Kinley et al. (2016a) concluded that the red algae _Asparagopsis_ spp. showed the most potential for CH$_4$ production decrease. Kinley et al. (2016b) further demonstrated that forage with the addition of 2% _Asparagopsis taxiformis_ could eliminate CH$_4$ production in vitro without negative effects on forage digestibility. In recent animal experiments, reduction of enteric CH$_4$ production by more than 98% was achieved with only 0.2% addition of freeze-dried and milled _Asparagopsis taxiformis_ to the to the organic matter (OM) content of feedlot cattle feed (Kinley et al., 2020).

Halogenated, biologically active secondary metabolites are pivotal in the reduction of CH$_4$ induced by _Asparagopsis_ (Abbott et al., 2020). Most of the reduction is ascribed to bromoform (CHBr$_3$) inhibition of the CH$_4$ biosynthetic pathway within methanogens (Machado et al., 2016). CHBr$_3$ as
a natural halogenated volatile organic compound originates from chemical and biological sources including marine phytoplankton and macroalgae (Carpenter et al., 2000; Quack and Wallace, 2003). When emitted to the atmosphere, CHBr$_3$ has an atmospheric lifetime shorter than six months and is often referred to as a very short-lived substance (VSLS). The halogenated VSLSs have drawn considerable interest because of their potential to deplete stratospheric ozone (Engel and Rigby, 2018). Bromoform is the dominant compound among bromine-containing VSLS emissions, resulting mostly from natural sources (Quack and Wallace, 2003) and to a lesser degree from anthropogenic production (Maas et al., 2019; 2021). With an atmospheric lifetime of about 17 days (Carpenter and Reimann, 2014), CHBr$_3$ can deliver bromine to the stratosphere under appropriate conditions of emission strength and vertical transport (e.g., Aschmann et al., 2009; Liang et al., 2010; Tegtmeier et al., 2015, 2020) and thus contribute to ozone depletion at middle and high altitudes (e.g., Yang et al., 2014; Sinnhuber and Meul, 2015). Global research on enabling large-scale seaweed Asparagopsis farming is increasing (Black et al., 2021) as it appears to be one of the most promising options as an antimethanogenic feed ingredient to achieve carbon neutrality in the livestock sector within the next decade (Kinley et al., 2020; Roque et al., 2021). In consequence, the environmental impact of CHBr$_3$ due to Asparagopsis farming also needs to be explored and elucidated.

The hypothesis was that large scale cultivation of Asparagopsis would not contribute significantly to depletion of the ozone layer. The aim of this study was elucidation of anthropogenic and natural processes that may contribute to CHBr$_3$ emissions inherent in large scale production of Asparagopsis spp. and the subsequent impact of CHBr$_3$ release to the atmosphere by using cultivation in Australia as the model. Specific objectives were to inform the industry on: (i) the potential impact of CHBr$_3$ associated with mass production of Asparagopsis on atmospheric halogen budgets and ozone depletion; (ii) potential impacts relative to variability in regional climate, atmospheric conditions, and convection trends with different potentials for transport of CHBr$_3$ to stratospheric ozone; (iii) the combined CHBr$_3$ emissions potential of ocean and terrestrial based cultivation of Asparagopsis to supply sufficient biomass for up to 50% of beef feedlot and dairy cattle in Australia; and (iv) extrapolation of the impacts of production to requirements on a global scale.

2. Data and Method
The potential impact of CHBr$_3$ on the atmospheric bromine budget and stratospheric ozone depletion, associated with Asparagopsis spp. mass production was assessed for assumed annual yields and particular production scenarios of macroalgae in Australia. Terrestrial systems cultivation and open ocean cultivation under different harvest conditions, variations of seaweed yield and growth rates for various scenarios and locations were tested as described in the following subsections.

2.1 Cultivation Scenarios

The cultivation scenarios in this study assume that sufficient seaweed is grown to supply Asparagopsis spp. to 50% of the Australian herds of beef cattle in feedlots (100%: ~1.0×10$^6$) and dairy cows (100%: ~1.5×10$^6$). For effective reduction of CH$_4$ production from ruminants, a 0.38% addition of freeze-dried and milled Asparagopsis taxiformis to the daily feed dry matter intake (DMI) is required (Kinley et al., 2020). This results in daily feed additions of 38 g dry weight (DW) Asparagopsis per head of feedlot cattle and 94 g DW Asparagopsis per head of dairy cows. In total, the required annual yield amounts to 3.4674×10$^4$ Mg DW Asparagopsis to supplement the feed of roughly 50% of the Australian feedlot cattle and Australian dairy cows. Assuming that fresh weight (FW) has a DW content of 15%, a total of 2.3116×10$^5$ Mg FW Asparagopsis needs to be harvested every year.

For a global scenario, we make the functional assumptions that: (i) there would be adoption of 30% of the global feed base to be supplemented with Asparagopsis farmed in Australia to reduce ruminant CH$_4$ production worldwide; (ii) Asparagopsis would be adopted by 50% of Australia’s feedlot and dairy industries; and (iii) this is approximately equivalent to 1% of the global feedlot and dairy herds for the purpose of both assumed magnitude of production and adoption relevant for calculations of supply and emissions. This export scenario requires for 30 times increased production compared to the Australian scenario if all the required Asparagopsis was to be cultivated in Australia and an annual harvest of ~1 Tg DW Asparagopsis would be needed from Australian waters.

For the future farm distributions in Australia, we assume that Asparagopsis will be cultivated in open ocean systems and terrestrial confinement systems (that may include, but not limited to, tanks, raceways, and ponds) located near Geraldton, Triabunna, and Yamba (Figure 1). We assume that
one third of the required annual yield (=1.1558 × 10^4 Mg DW) is grown near Triabunna (T), with
60% in terrestrial systems and 40% in open ocean farms, one third is grown in terrestrial systems
at Yamba (Y), and the last third is grown in the open ocean in Geraldton (G). For comparison of
the environmental impact, we also adopt a tropical scenario where all farms with their total annual
yield of 3.4674 × 10^4 Mg DW are assumed to be situated near Darwin.

The emissions of CHBr₃ from the macroalgae farms can be derived based on estimates of the
standing stock biomass. For any given farming scenario, the standing stock biomass \( B_f \) (g DW) is
a function of time \( t \) and can be calculated from the initial biomass \( B_i \) (g DW) and the specific
growth rate \( GR \) (%/day) according to Hung et al. (2009):

\[
B_f(t) = B_i \cdot (1 + GR/100)^t
\]  

Terrestrial systems and open ocean cultivation scenarios are assuming a fixed targeted annual yield.
For a given initial biomass and growth rate, the length and frequency of the growth periods per
year need to be chosen accordingly, to achieve the required final yield. Yong et al., (2013) checked
the reliability of different equations for seaweed growth rate determination by comparing the daily
seaweed weight cultivated under optimized growth condition, and the most reliable relationship
between initial and final weight leads to the form of Eq (1). We also applied several growth rates
from 1 to 10% to show the possible influence of this parameter on the overall emissions of the
algae. Average growth rates of *Asparagopsis* ranged from 7 to 13 %/day in samples from tropical
and sub-tropical Australia during short-term experiments (Mata et al., 2017). We used a lower
growth rate of 5% for our scenario to provide an upper estimate of potential CHBr₃ emissions.
Note that emissions decrease by 27% when using a growth rate of 7% as demonstrated in section
3.1.

Figure 2 provides an example of the variations of standing stock of *Asparagopsis* for the farms of
Geraldton (all open ocean) and Yamba (all terrestrial systems) with a growth rate of 5% per day.
For the open ocean cultures, we assume a scenario of six harvests per year and 60 day growth
periods to obtain the annual yield (Elsom, 2020). For a sensitivity study, we assume an alternative
scenario based on the same initial biomass, but only one harvest per year. As evident from Figure
2, the same annual yield can be achieved with one harvest per year if applying an extended growth
period of 96 days. For the tank cultures, a harvest every 5 days (73 harvests per year) is assumed
as a realistic scenario (Elsom, 2020).
Figure 1. Locations of actual and theoretical *Asparagopsis* farms in Geraldton, Triabunna, Yamba, and Darwin.
Figure 2. Standing stock biomass of *Asparagopsis* cultivation a) in the open ocean for a 60-day growth period and 96 day growth period and b) in terrestrial systems culture for a 5 day growth period. Each of the three scenarios will achieve an annual yield of $1.1558 \times 10^4$ Mg DW.

2.2 *Asparagopsis* CHBr$_3$ release rates

Rates of the CHBr$_3$ content in *Asparagopsis* given in the literature range between 3.4 to 43 mg CHBr$_3$/g DW, with values around 10 mg CHBr$_3$/g DW appearing to be realistic in current cultivation (Burreson and Moore, 1976; Mata et al., 2012, 2017; Paul et al., 2006; Vucko et al., 2017). We assume that *Asparagopsis* strain selection cultivated for feed supplements will lead to high yielding CHBr$_3$ varieties thus we assume augmented CHBr$_3$ production with a mean content...
of 21.7 mg CHBr$_3$/g DW (Magnusson et al., 2019) for this study. Very few values on the CHBr$_3$ release from *Asparagopsis* have been reported in the literature. A constant release of 1100 ng CHBr$_3$/g DW hr$^{-1}$ was measured for *Asparagopsis armata* tetrasporophyte, which has a CHBr$_3$ content of 14.5 mg CHBr$_3$/g DW (Paul et al., 2006). We assume a linear scaling between the CHBr$_3$ release rates and the content. Thus, a cultivated *Asparagopsis* for which we assume 21.7 mg CHBr$_3$/g DW should release around 1646 ng CHBr$_3$/g DW hr$^{-1}$, a rate which has been confirmed by Marshall et al. (1999). Therefore, for our calculations, we assume a constant release of 1600 ng CHBr$_3$/g DW hr$^{-1}$ for farmed *Asparagopsis* with a CHBr$_3$ content of 21.7 mg CHBr$_3$/g DW. These content and release rates are higher than for wild stock algae (Leedham et al., 2013; Nightingale et al., 1995) as the farming aims at high yielding CHBr$_3$ varieties. As available information on this topic is very sparse no variations of the release rate with life-cycle stages, season, location, or other environmental parameters was used in this study. Also, the two species *Asparagopsis armata* and *Asparagopsis taxiformis* were treated the same way as *Asparagopsis spp.*, as variations in CHBr$_3$ content and release within or between species are currently unknown (Mata et al., 2017) and more research on this topic is needed.

### 2.3 Parameterization of CHBr$_3$ Emission

The emissions of CHBr$_3$ from farmed macroalgae are a function of the standing stock biomass (in g DW) and can be calculated with the constant release rate ($R_{CHBr_3}$) of 1600 ng CHBr$_3$/g DW hr$^{-1}$ multiplied with the standing stock. The total release of CHBr$_3$ ($E_{CHBr_3}$) over the complete growth period of T days is given by the integral over the daily emissions from day 1 to day T:

$$E_{CHBr_3} = \int_{0}^{T} 24 \cdot B_i \cdot (1 + GR)^t \cdot R_{CHBr_3} dt = 24 \cdot B_i \cdot R_{CHBr_3} \left[\frac{(1+GR)^T-1}{\ln(1+GR)}\right]$$

(2)

For our atmospheric impact studies we assume, that all CHBr$_3$ released from the algae is emitted into the atmosphere at its location of production. An increasing seawater concentration of CHBr$_3$ shifts the equilibrium conditions between seawater and air towards the atmosphere, as CHBr$_3$ easily volatilizes to the atmosphere. Consequently, air-sea exchange acts as a relatively fast loss process for CHBr$_3$ in surface water. Oceanic sinks can also impact CHBr$_3$, but act on relatively long timescales. Degradation through halide substitution and hydrolysis results in the ocean sink CHBr$_3$ half-life of 4.37 years (Hense and Quack, 2009). Thus, most of the CHBr$_3$ contained in
Surface seawater is instantly outgassed into the atmosphere without oceanic loss processes playing a role as confirmed by the modelling study of Maas et al. (2020).

The air-sea exchange of CHBr₃ is expressed as the product of its transfer coefficient (kₐ) and the concentration gradient (∆c) (Eq. (3)). The gradient is between the water concentration (cₜ) and theoretical equilibrium water concentration (cₜ atm/H), where cₜ atm is the atmospheric concentration and H is Henry’s law constant (Moore et al., 1995a; Moore et al., 1995b).

\[ F = kₐ \cdot \Delta c = kₐ \cdot (cₜ - cₜ atm/H) \]  

The compound-specific transfer coefficient (kₐ) is determined using the air-sea gas exchange parameterization of Nightingale et al. (2000) (Eq. (4))

\[ kₐ = k \cdot \sqrt{Sc} / 660 \]  

The transfer coefficient k is a function of the wind speed at 10 m height (u₁₀): \( k = 0.2u₁₀^2 + 0.3u₁₀ \), and the Schmidt number (Sc) is a function of sea surface temperature (SST) from Quack and Wallace (2003), which is expressed as \( Sc = 4662.8 - 319.45 \cdot SST + 9.9012 \cdot SST^2 + 0.1159 \cdot SST^3 \).

In this study, we use the CHBr₃ sea-to-air flux climatology from Ziska et al. (2013) as marine background emissions. The global emission scenario from Ziska et al. (2013) is a bottom-up estimate of the oceanic CHBr₃ fluxes, generated from atmospheric and oceanic surface ship-borne \textit{in situ} measurements between 1979 to 2013. Due to the paucity of data the 35 year mean gridded data set was filled by inter- and extrapolating the \textit{in situ} measurement data. The oceanic emissions were calculated with the transfer coefficient parameterization of Nightingale et al. (2000) and 6-hourly meteorological data, which allow a temporal emission variability related to wind and temperature.

### 2.4 FLEXPART

To quantify the atmospheric impact of CHBr₃ emissions from macroalgae farming, the Lagrangian particle dispersion model FLEXPART (Pisso et al., 2019) is used. FLEXPART has been evaluated extensively in previous studies (e.g., Stohl et al., 1998; Stohl and Trickl, 1999). The model includes moist convection and turbulence parameterizations in the atmospheric boundary layer and free troposphere (Forster et al., 2007; Stohl and Thomson, 1999). The European Centre for Medium-Range Weather Forecasts (ECMWF) reanalysis product ERA-Interim (Dee et al., 2011) with a
horizontal resolution of 1° x 1° and 60 vertical model levels is used for the meteorological input fields, providing air temperature, winds, boundary layer height, specific humidity, as well as convective and large-scale precipitation with a 3-hour temporal resolution.

We conduct FLEXPART simulations for different emission scenarios as explained in the following and summarized in Table 1:

1.) Australian scenarios: CHBr₃ emissions from the *Asparagopsis* farming in Geraldton, Triabunna, and Yamba are calculated for an overall annual yield of 3.4674×10⁴ Mg DW according to Equation 2.

For the terrestrial systems, 5 day growth periods are assumed resulting in 73 harvests per year. For the open ocean, the assumption of different growth periods results in three sub-scenarios a) 6 times 60 day growth periods with the first period starting on January 1st (referred to as GTY_O60), b) one 96 day growth period starting on January 1st (GTY_O96_Jan), and c) and another starting on July 1st (GTY_O96_Jul).

For the last Australian scenario, we assume that all farms are located around Darwin in the Northern Territory tropics with 6 times 60 day growth periods in the open ocean and 73 times 5 day growth periods in the terrestrial systems (Darwin_O60). While this is an unlikely scenario according to current plans, it is useful to demonstrate the influence of potential farming locations on their environmental impact.

2.) Global scenarios: Emissions from *Asparagopsis* farming in Geraldton, Triabunna, and Yamba are estimated according to the annual yield, upscaled by a factor of 30 to global requirements, amounting to 1.04×10⁶ Mg DW. Growth periods and harvesting frequencies are set up in the same way as for the Australian scenarios. Short names of the global scenarios are the same as for the Australian scenarios with the additional label 30x.

3) Background scenario: Emission from Ziska et al. (2013) for the entire coastal region around Australia defined as all 1°x1° grid cells directly neighbouring the coastline (Ziska_Coast).

4.) Extreme event scenarios: We assume extreme conditions where a hypothetical tropical cyclone causes implausible release of all CHBr₃ from the macroalgae farm and water into the atmosphere. We focus on the case study of Geraldton and the tropical cyclone Joyce, which occurred from 6-13 January 2018 around western Australia. We base the amount of available macroalgae biomass on the Australian scenario and assume that the entire CHBr₃ content of all *Asparagopsis* at this location is released at once. The two scenarios defined here assume that the tropical cyclone occurs at the end of the 60 day growth period (Geraldton_Ex60) resulting in the release of 41.8 Mg CHBr₃.
(21.7 mg CHBr\textsubscript{3}/g DW * 1926 Mg DW) or at the end of the 96 day growth period (Geraldton_Ex96) resulting in the release of 250.8 Mg CHBr\textsubscript{3} (21.7 mg CHBr\textsubscript{3}/g DW* 1.1558×10\textsuperscript{4} Mg DW).

The daily model output is recorded for all simulations. For the extreme event, which assumes the destruction of a farm (Geraldton-Ex), the 3 hourly output is recorded. For all simulations, except the background scenario and extreme scenario, trajectories are released from four regions of the size of: a) Geraldton (open ocean, 11558 ha): 0.1°x0.1°; b) Triabunna (open ocean, 4623 ha): 0.06°x0.06°; c) Triabunna (terrestrial systems, 126 ha): 0.01°x0.01°; and d) Yamba (terrestrial systems, 210 ha): 0.01°x0.01°. For the tropical and extreme scenarios, trajectories are released from the Darwin and Geraldton farms, respectively. For the background scenario Ziska_Coast, trajectories are released from the 1.0°x1.0° grid along the Australian coastline. The amount of released CHBr\textsubscript{3} is evenly distributed among the trajectories and is depleted during the Lagrangian simulations according to the atmospheric half-life of 17 days (e-folding lifetime of 24 days) (Hossaini et al., 2010; Montzka and Reimann, 2010).
Table 1. Detailed information on the scenarios set up for the atmospheric transport simulations with FLEXPART (Geraldton, Triubanna, and Yamba: GTY)

<table>
<thead>
<tr>
<th>Name</th>
<th>Total Yield (Mg DW)</th>
<th>CHBr$_3$ Emissions (Mg)</th>
<th>Notes</th>
<th>Simulation Period</th>
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</thead>
<tbody>
<tr>
<td><strong>Australian Scenarios</strong></td>
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<tr>
<td>GTY_O60</td>
<td>Total: 34674</td>
<td>Total: 27.3</td>
<td>6 harvests (every 60 days) in the open ocean; 73 harvests (every 5 days) in the terrestrial systems.</td>
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<td>Open Ocean:</td>
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<td>G: 11558</td>
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<td>T: 4623</td>
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<td>Y: -</td>
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<td></td>
<td>Terrestrial systems:</td>
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<td>G: -</td>
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<td>Y: 11558</td>
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<tr>
<td>GTY_O96_Jan</td>
<td>Terrestrial systems:</td>
<td></td>
<td>1 harvest (after 96 days) in open ocean; 73 harvests (every 5 days) in terrestrial systems. Growth in open ocean starts from 01.01.2018.</td>
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<td>G: -</td>
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<td>Y: 11558</td>
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<tr>
<td>GTY_O96_Jul</td>
<td>Terrestrial systems:</td>
<td></td>
<td>Same as GTY_O96_Jan but with growth in open ocean starting from 01.07.2018.</td>
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<td>Y: 11558</td>
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<tr>
<td>Darwin_O60</td>
<td>Total: 34674</td>
<td>Total: 27.3</td>
<td>Same as GTY_O60 but with farms near Darwin</td>
<td>01.01.2018 - 31.12.2018</td>
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<td>Open Ocean:</td>
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<td>Darwin: 16181</td>
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<td>Terrestrial systems:</td>
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<td>Darwin: 18493</td>
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<td>GTY_O60_30x</td>
<td>Total: 1.04022×10$^6$</td>
<td>Total: 819</td>
<td>Same as GTY_O60 but with initial biomass and areas 30 times larger.</td>
<td>2-month spin-up</td>
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<td>Open Ocean G: 3.4674×10$^3$</td>
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<td>T: 1.3869×10$^6$</td>
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<td>Terrestrial systems:</td>
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<td>T: 2.0805×10$^5$</td>
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<td>Y: 3.4674×10$^3$</td>
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<tr>
<td>GTY_O96_Jan_30x</td>
<td>Terrestrial systems:</td>
<td></td>
<td>Same as GTY_O96_Jan but with initial biomass and areas 30 times larger.</td>
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<td>G: -</td>
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<td>T: 1.3869×10$^6$</td>
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<td>T: 2.0805×10$^5$</td>
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<td>Y: 3.4674×10$^3$</td>
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<tr>
<td>GTY_O96_Jul_30x</td>
<td>Terrestrial systems:</td>
<td></td>
<td>Same as GTY_O96_Jul but with initial biomass and areas 30 times larger.</td>
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<td>G: -</td>
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<td>T: 1.3869×10$^6$</td>
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<td>Terrestrial systems:</td>
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<td>T: 2.0805×10$^5$</td>
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<td></td>
<td>Y: 3.4674×10$^3$</td>
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<tr>
<td>Darwin_O60_30x</td>
<td>Total: 1.04022×10$^6$</td>
<td>Total: 819</td>
<td>Same as Darwin_O60 but with initial biomass and areas 30 times larger.</td>
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<td>Open Ocean Darwin:</td>
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<td>4.8543×10$^3$</td>
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<td>Darwin: 438</td>
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The ozone depletion potential (ODP) is defined as the time-integrated potential destructive effect of a substance to the ozone layer relative to that of the reference substance CFC-11 (Wuebbles, 1983). The ODP is a well-established and extensively used concept traditionally defined for anthropogenic long-lived halogens. However, the concept has been also applied to VSLSs (Brioude et al., 2010; Pisso et al., 2010): unlike the ODP for long-lived halocarbons, which is one constant number, the ODP of a VSLS is a function of time and location of the emissions. This variable number still describes the time-integrated ozone depletion resulting from a CHBr\(_3\) unit mass emission relative to the ozone depletion resulting from a unit mass emission of CFC-11. However, the trajectory-derived ODP of e.g. CHBr\(_3\) is calculated as a function of location and time of the potential emissions. As for the classical ODP, and independently of the total amount of CHBr3 emitted, the time and space dependent ODP describes only its potential but not the actual damaging effect to the ozone layer. The fraction of originally emitted VSLSs reaching the stratosphere depends strongly on the meteorological conditions. In particular, it shows a pronounced seasonality. Here we apply ODP values adapted from Pisso et al. (2010), originally calculated for a VSLS with a lifetime very similar to that of CHBr\(_3\). 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. The former is uniform for all species modelled and is based on the calculation of the expected stratospheric residence time of a Lagrangian particle entering the stratosphere. The latter is based on the probability of stratospheric
injection of a given unit emission of the tracer at the ground. The probability of injection depends not only on the fraction of air reaching the tropopause but also on the time the air mass takes from the ground to the tropopause. This is because during the transit of the air mass through the troposphere, the precursor is chemically degraded, and the solubility of the products leads to mass loss due to wet deposition.

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 CHBr$_3$ emissions with the trajectory-derived ODP at each grid point. The ODP-weighted emissions provide insight in where and when CHBr$_3$ is emitted that impacts stratospheric ozone (Tegtmeier et al., 2015). The absolute values are subject to relatively large uncertainties arising from uncertainties in the parameterization of the convective transport. Furthermore, the here applied ODP values do not consider product gas entrainment and provide therefore a lower limit of the impact of CHBr$_3$ on stratospheric ozone. Taking into account product gas entrainment can lead to 30% higher ODP values (Engel and Rigby, 2018; Tegtmeier et al., 2020), but has no large impact on the here presented comparison of global ODP-weighted CHBr$_3$ emissions with farm-based ODP-weighted CHBr$_3$ emissions.

3. CHBr$_3$ Emission and Atmospheric Mixing Ratio

3.1 CHBr$_3$ Emissions

As shown in Eq. (2), the total CHBr$_3$ emissions are determined by the growth rate, growth period and initial biomass. For our scenarios based on selected fixed growth rates, the growth periods are adjusted so that the intended annual yield (\(3.5 \times 10^4\) Mg DW) is achieved. We conduct a sensitivity study to analyze how much the total emissions change for variations of the length and number of the growth periods for a fixed annual yield. For this purpose, we compare Geraldton farming for GTY_O60 (open ocean, six 60 day growth periods) with Geraldton farming for GTY_O96 (open ocean, one 96 day growth period) and Yamba farming for GTY_O60 (terrestrial systems, 73 growth periods of 5 days). Our estimates reveal that the annual release of CHBr$_3$ from Asparagopsis is the same for all three case studies (Fig. 3a), indicating that for a fixed annual yield and growth rate, the culture conditions of open ocean and tank farming are not important for VSLSs emissions.
A second sensitivity study investigates the variations of CHBr₃ emissions for different growth rates and the same fixed annual yield. For this purpose, we compare Geraldton farming (open ocean, with an intended annual yield of \(\sim 1.1 \times 10^4\) Mg DW) for different growth rates varying between 1% and 10%. The scenario with a 5% growth rate corresponds to Geraldton farming for GTY_O60 (open ocean, six 60 day growth periods), while for the other growth rates the growth periods have been adjusted to achieve the same annual yield.

The CHBr₃ emissions depend strongly on the growth rates (Fig 3b), with emission calculated for a 1% growth rate being almost 10 times higher than the emissions calculated for a 10% growth rate. For a lower growth rate, the initial biomass needs to be higher to achieve the targeted seaweed yield (\(\sim 1.1 \times 10^4\) Mg) after one year and/or the growth period needs to be longer, thus resulting in larger amounts of biomass in the ocean and higher annual CHBr₃ emissions. Vice versa, for higher growth rates, the annual oceanic biomass is smaller and total emissions are lower.

The overall emissions from the intended Australian seaweed farming of \(\sim 3.5 \times 10^4\) Mg DW range from 13.5 Mg (0.05 Mmol) for a 10% growth rate to 134 Mg (0.5 Mmol) per year for a 1% growth rate. For the growth rates higher than 5%, the differences of CHBr₃ emissions are less significant than those derived for the lower growth rates. In our study, we choose 5% growth rate as representative, which leads to emissions of \(\sim 27\) Mg (0.1 Mmol) CHBr₃ per year for the targeted final yield. For the global scenario with an annual yield of \(\sim 1.0 \times 10^6\) Mg DW (30 times of the Australian target), the emissions would range from 412 Mg (1.6 Mmol) to 4014 Mg (16 Mmol) per year, with the annual emission of 810 Mg (3.2 Mmol) for 5% growth rates.

Interestingly, the potential local emissions for all the farming scenarios are generally 3 to 6 orders of magnitude higher than the background coastal emissions. The maximum climatological emissions derived from available observations (Ziska_Coast) are around 2000 pmol m⁻² hr⁻¹ for the coastal waters of Australia, while the emissions from an Asparagopsis farm can reach more than 2.0 \(\times 10^4\) pmol (2 μmol) m⁻² hr⁻¹ from a terrestrial system and more than 5.0 \(\times 10^5\) pmol m⁻² hr⁻¹ from the open ocean. These differences are to a large degree related to the fact that the Ziska_Coast is given on a 1.0°x1.0° grid, with high coastal values averaging out over the relatively wide grid cells, while the values derived for the farms apply to much smaller areas. Tank emission rates (0.01°x0.01°) and open ocean farming emission rates (0.1°x0.1°) averaged over a 1°x1° grid cell result in 200 pmol CHBr₃ m⁻² hr⁻¹ and 5000 pmol CHBr₃ m⁻² hr⁻¹, respectively, thus being very similar to the Ziska emissions.
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, both with a total annual yield of 1.1558$\times$10$^4$ Mg DW.
We use the CHBr$_3$ emissions calculated in section 3.1 to simulate the atmospheric CHBr$_3$ mixing ratios for each Asparagopsis farming scenario. Background CHBr$_3$ levels are calculated based on the Ziska et al. (2013) Australian coastal emissions (Ziska_Coast). The temporal evolution of CHBr$_3$ mixing ratio with height shows that the CHBr$_3$ resulting from the Australian farming scenarios are negligible (see Figure S1) compared to the coastal background emissions of Australia (Ziska_Coast).

However, for the global scenarios (Figure 4), atmospheric CHBr$_3$ is comparable to CHBr$_3$ resulting from Australian coastal background emissions, especially near the end of the growth period in the open ocean. For almost all scenarios (except for GTY_O96_Jul_30x), the emissions generally reach higher into the atmosphere in the first three months of the year with enhanced values around 15 km, reflecting the stronger convection during austral summer. For open ocean emissions occurring during late austral winter (GTY_O96_Jul_30x, Figure 4c), high CHBr$_3$ mixing ratios are found around September, however at a lower altitude range compared to the equivalent scenario with open ocean emission occurring during late austral summer (GTY_O96_Jan_30x; Figure 3b).

The spatial distribution of annual mean CHBr$_3$ at 1 km (Figure 5) further confirms the insignificance of the signals from the Australian farming scenarios compared with the background CHBr$_3$ values. For the global scenarios, localized regions of high mixing ratios are found near the locations of the farms due to the stronger emission. For Darwin_O60_30x, the belt of high mixing ratios is extending northwestward, due to the prevailing easterlies in the tropics. At higher altitudes (e.g., 5 km and 15 km; Figure S2-S3), localized high CHBr$_3$ is only found near Darwin for the Darwin_O60_30x scenario, reflecting that strong tropical convection is needed to transport short lived gases to such altitudes.

The results above suggest that in the boundary layer, global scenarios and extreme events could lead to CHBr$_3$ comparable mixing ratios as those from the background scenario. Only in the global tropical scenario (Darwin_O60_30x), CHBr$_3$ mixing ratios, which are larger than the background values, can be found at high altitudes (Figure 4).

Simulations of the two extreme scenarios (Geraldton_Ex) for 60 and 96 day growth periods are shown in Figure 6. For the Geraldton_Ex simulations, we assume the implausible scenario that
cyclone Joyce could destroy the farm on the day of harvest in January and the total CHBr$_3$ content of the *Asparagopsis* stock was simultaneously released to the atmosphere during the event. Both scenarios lead to significant CHBr$_3$ mixing ratios in the atmosphere, especially at altitudes below 5 km. Among the two scenarios, the Geraldton_Ex96 contributes the larger amount of CHBr$_3$ emission, as the macroalgae experienced a longer growth period, so the biomass was higher and had accumulated more CHBr$_3$. When averaged over the same period (Jan 9-Jan 26, 2018), the CHBr$_3$ mixing ratios from Geraldton_Ex96 are much larger than those from Ziska_Coast (Figure 6) below 5 km, and signals with comparable magnitudes are found at 15 km. As mentioned in section 3.1, the local CHBr$_3$ emissions due to the seaweed cultivation are generally higher than coastal emission given on 1.0x1.0 grid. However, due to the relatively small spatial extent of the farms, the emissions quickly dilute in the atmosphere, and the magnitude of the mixing ratios decline rapidly off the coast and vertically.
Figure 4. Altitude-time cross-sections of CHBr$_3$ averaged over [10°-45° S, 105°-165° E] of CHBr$_3$ mixing ratio from a) GTY_O60_30x, b) GTY_O96_Jan_30x, c) GTY_O96_Jul_30x, d) Darwin_O60_30x, and e) Ziska_Coast.

Figure 5. Annual mean CHBr$_3$ spatial distribution from GTY_O60, GTY_O60_30x, GTY_O96_Jan, GTY_O96_Jan_30x, GTY_O96_Jul, GTY_O96_Jul_30x, Darwin_O60, Darwin_O60_30x, and Ziska_Coast at 1 km altitude.
Figure 6. 17-day average of spatial distribution and altitude-time cross-sections of CHBr$_3$ mixing ratio averaged over [10°-45° S, 105°-165° E] for Geraldton_Ex60, Geraldton_Ex96, and Ziska_Coast.

4. Ozone depletion potential for CHBr$_3$

The ODP distribution for the region around Australia, South-East Asia, and the Indian Ocean for the Southern Hemisphere (SH) summer and winter is shown in Figure 7. The ODP distribution changes strongly with season as the transport of short-lived halogenated substances such as CHBr$_3$ depends on the seasonal variations of the location of the Intertropical Convergence Zone (ITCZ). Highest ODP values of 0.5, which imply that any amount (per mass) of CHBr$_3$ released from the specific location will destroy half as much stratospheric ozone as the same amount of CFC-11 released from this location, are found during July over the Maritime continent and during January over the West Pacific south of the equator. The northern Australian coastline shows highest ODP
values during January when the thermal equator and the ITCZ are shifted southwards and ODP values for Yamba and Darwin are 0.26 and 0.29, respectively. The other two locations as well as all four locations during SH winter, show ODP values of only up to 0.1. As demonstrated in section 3, the total annual CHBr$_3$ emissions from any location are independent of the details of the farming practice, however, the ODP-weighted emissions change for the different scenarios as the growth periods fall into different seasons with varying ODP values. In general, the scenario of one harvest period in SH summer leads to larger ODP-weighted emissions when compared to the same biomass harvested throughout the year. In addition to the harvesting practice, the locations of the farms have a large impact on the efficiency of the CHBr$_3$ transport to the stratosphere and thus on the ODP-weighted emissions. The ODP-weighted emissions of CHBr$_3$ for different emission scenarios are shown in Figure 8. Asparagopsis farming at GTY (GTY_O60) leads to additional CHBr$_3$ emissions of up to 2.53 Mg per year. If all farming (~3.5×10$^4$ Mg DW Asparagopsis) occurs in Darwin (Darwin_O60), ODP-weighted emissions would increase to 6.48 Mg CHBr$_3$ per year. In comparison, all naturally occurring emissions around the Australian coastline (Ziska_coast) lead to OPD-weighted CHBr$_3$ emissions of 221.52 Mg per year. In consequence, Asparagopsis farming in the three locations Geraldton, Triabunna and Yamba would lead to an increase of the ODP-weighted emissions from Australian coastal emissions of 1.14%. If all farming would take place in Darwin, ODP-weighted CHBr$_3$ emissions would increase by 2.93%.

As the global ODP-weighted emissions were estimated to be around 4.0×10$^4$ Mg per year (Tegtmeier et al., 2015), the additional contribution due to the Australian farming scenarios in GTY or Darwin would be negligible increasing the contribution of CHBr$_3$ emissions to ozone depletion by 0.006% and 0.016%, respectively. Even if the farming would be upscaled to cover the global needs (~1.0×10$^6$ Mg DW), the ODP-weighted CHBr$_3$ emissions would only increase to 75 Mg and 195 Mg for farming in GTY (GTY_O60_30x) and Darwin (Darwin_O60_30x), respectively. Thus produced CHBr$_3$ would increase the current contribution of CHBr$_3$ to stratospheric ozone depletion by 0.19% and 0.48%, which is again a very small contribution. To assess the increase of the ODP-weighted CHBr$_3$ emissions under the most extreme and implausible conditions, we envision the total harvest of one year, which contains 752 Mg (21.7 mg CHBr$_3$/g DW*3.4674×10$^4$ Mg DW) CHBr$_3$, stored in a warehouse of 50 x 25 x 5 m in either of the four locations. We assume that the facility is destroyed, and all 750 Mg released to the
atmosphere. Then maximum ODP-weighted CHBr$_3$ emissions would occur for the release in Darwin during January and amount to 215.9 Mg almost doubling the ODP-weighted coastal CHBr$_3$ emissions of Australia. If the entire content of $\approx 1.0 \times 10^6$ Mg $Asparagopsis$ DW (21.7 mg CHBr$_3$/g DW*1.04022$\times 10^6$ Mg DW=2.2573$\times 10^4$ Mg CHBr$_3$) would be released in Darwin, the additional contribution of CHBr$_3$ to global ozone depletion could reach 16%. 

Figure 7. Spatial distribution of the ODP in January and July.
5. Summary and Conclusions

In this study, we assessed the potential risks of CHBr$_3$ released from *Asparagopsis* farming near Australia for the stratospheric ozone layer by analyzing different cultivation scenarios. We conclude that the intended operation of *Asparagopsis* seaweed cultivation farms with an annual yield of 3.4674×10$^4$ Mg DW in either open ocean or terrestrial cultures at the locations Triubanna, Yamba, Geraldton, and Darwin will not impact the ozone layer under normal operating conditions. For Australia scenarios with an annual yield of ~3.5×10$^4$ Mg DW and algae growth rate of 5% per day, the expected annual CHBr3 emission from the considered *Asparagopsis* farms into the atmosphere (~27 Mg, 0.11 Mmol) is less than 0.9% of the coastal Australian emissions (~3109 Mg, 12.3 Mmol). This contribution is negligible from a global perspective by adding less than 0.01% to the worldwide CHBr3 emissions from natural and anthropogenic sources. The overall emissions from the farms would be even smaller with a faster growth rate for the same annual yield. We have assumed a high CHBr$_3$ production of 21.7 mg/g DW from superior strains and
expected lower CHBr$_3$ production of 14 mg/g DW would likewise reduce emissions to the atmosphere.

The local CHBr$_3$ emissions from the *Asparagopsis* farms could be larger than emissions from coastal Australia. However, the overall atmospheric impact of the *Asparagopsis* farms is negligible, as the CHBr$_3$ dilutes rapidly and degrades in the atmosphere under normal weather conditions.

Mixing ratios of CHBr$_3$ are generally dominated by the coastal Australian emissions. In global scenarios with annual yield $\sim 1.0 \times 10^6$ Mg DW, localized CHBr$_3$ mixing ratios comparable to the background values can be found in the lower troposphere. In the upper troposphere, on the other hand, mixing ratios larger than background values only appear in the global tropical scenario (Darwin_O60_30x). The release of the complete CHBr$_3$ content from the macroalgae to the environment on very short timescales (e.g., days) due to extreme weather situations could contribute significant amounts to the atmosphere, especially during times when the standing stock biomass is relatively large (Geraldton_Ex96). While such extreme scenarios could lead to much larger mixing ratios than background values, such mass release events are implausible because even if a farm was totally destroyed the seaweed stock could not instantaneously release all the accumulated CHBr$_3$. Such scenarios have been included here to evaluate a catastrophic and likely impossible worst-case scenario.

The impact of CHBr$_3$ from the proposed seaweed farms on the stratospheric ozone layer is assessed by weighting the emissions with the ozone depletion potential of CHBr$_3$. In total, Australia scenarios could lead to additional ODP-weighted CHBr$_3$ emissions of up to 2.53 Mg per year with farms located in Geraldton, Triubana and Yamba. With all farming performed in Darwin (Darwin_O60), the emitted CHBr$_3$ could reach the stratosphere on shorter time scales and ODP-weighted emissions would increase to 6.48 Mg, which is less than 0.016% of the global ODP-weighted emissions. For global tropical scenario (Darwin_O60_30x), the ODP-weighted emissions amount to 175 Mg, increasing the global ozone depletion by 0.48%, resulting in a very small contribution.

We note that all data characterizing the potential systems for the production of *Asparagopsis* are based on few available literature data, lab scale tests and relatively small-scale field trials. This not only places limitations on the technological representativeness of a future system and the temporal validity of the study, but also demonstrates importance for directed studies, especially on the release of CHBr$_3$ from *Asparagopsis* during cultivation. As this understanding evolves so will the
cultivation and processing technologies engineered to conserve the antimethanogenic CHBr$_3$ in the seaweed biomass which is the primary value feature of *Asparagopsis*. These limitations are largely mitigated in our study by evaluating various environmental and meteorological conditions ranging from conservative to most extreme scenarios and by investigating different farming practices based on various sensitivity studies.

Data availability

The CHBr$_3$ emission data and FLEXPART output can be obtained from the authors on request via BQ (bquack@geomar.de), ST (susann.tegtmeier@usask.ca), or YJ (yue.jia@noaa.gov).

Author Contributions

BQ initialized the idea. YJ, BQ, and ST carried out the calculations and analysis. YJ performed the FLEXPART simulations and produced the figures. YJ, BQ, and ST wrote the manuscript with the contribution from other co-authors RK and IP. RK contributed to conceptualization, design, writing, editing, procurement of funding. All the authors contributed to discussions and revisions of the manuscript.

Competing interests

The authors declare that they have no conflict of interest.

Acknowledgements

The authors wish to acknowledge CSIRO, FutureFeed, and Sea Forest for their provision of technical knowledge, data, and insight into *Asparagopsis* supply chains in Australia. The authors would like to thank the European Centre for Medium-Range Weather Forecasts (ECMWF) for the ERA-Interim reanalysis data and the FLEXPART development team for the Lagrangian particle dispersion model used in this publication. The FLEXPART simulations were performed on resources provided by the University of Saskatchewan.
Reference


