



1 **How Marine Emissions of Bromoform Impact the**

2 **Remote Atmosphere**

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10 **Abstract**

11 Oceanic emissions of very short lived halocarbons (VSLH), such as CHBr_3 , are important for the halogen
12 budget of the atmosphere. It is an open question how localized elevated emissions in coastal and
13 upwelling regions and low background emissions, typically found over the open ocean, impact the
14 atmospheric VSLH distribution. In this study, we use the Lagrangian dispersion model FLEXPART to
15 simulate atmospheric CHBr_3 resulting from uniform background emissions, on the one hand, and from
16 elevated emissions observed during three tropical cruise campaigns, on the other hand.

17 The simulations demonstrate that the atmospheric CHBr_3 distributions due to uniform background
18 emissions are highly variable with accumulations taking place in regions of low wind speed. This relation
19 holds on regional and global scales demonstrating the importance of the atmospheric transport for the
20 distribution of short-lived trace gases with lifetimes in the range of days to weeks.

21 The impact of localized elevated emissions, measured during three research cruises, on the atmospheric
22 CHBr_3 distribution varies significantly from campaign to campaign. The estimated impact depends on
23 the strength of the emissions and the meteorological conditions. In the open waters of the western Pacific
24 and Indian Ocean, localized elevated emissions only slightly increases the background concentrations of
25 atmospheric CHBr_3 , even when 1° wide source regions along the cruise tracks are assumed. Near the
26 coast, elevated emissions, including hotspots up to 100 times larger than the uniform background
27 emissions, can be strong enough to be distinguished from the atmospheric background. However, it is
28 not necessarily the highest hotspot emission that produces the largest enhancement, since the tug-of-war
29 between fast advective transport and local accumulation at the time of emission is also important.

30 Our analyses contribute to a better understanding and prediction of the timing and regional characteristics
31 of tropospheric CHBr_3 distribution. Significantly, our results demonstrate that transport variations of the
32 atmosphere itself are sufficient to produce highly variable VSLH distributions, and elevated VSLH in
33 the atmosphere do not always reflect a strong localized source. Localized elevated emissions can be
34 obliterated by the highly variable atmospheric background, even if they are orders of magnitude larger
35 than the average open ocean emissions.

36



37 1. Introduction

38

39 Very short lived halocarbons (VSLHs) with atmospheric lifetimes shorter than 6 months have natural
40 oceanic sources which are dominated by brominated and iodinated compounds (Carpenter and Liss, 2000;
41 Quack et al., 2004; Law et al., 2006). VSLHs have drawn considerable interest due to their contribution
42 to stratospheric ozone depletion and tropospheric chemistry (Solomon et al., 1994; Dvortsov et al., 1999;
43 Salawitch et al., 2005; Feng et al., 2007; Tegtmeier et al., 2015; Hossaini et al., 2015). In this work, we
44 focus on the VSLH bromoform (CHBr_3), since most organic oceanic bromine is released into the
45 atmosphere in this form.

46 CHBr_3 concentrations measured in ocean waters are characterized by large spatial variability with
47 elevated abundances in phytoplankton blooms (Baker et al., 2000; Liu et al., 2013) and equatorial and
48 upwelling regions due to biological sources (Carpenter et al., 2009; Quack and Wallace, 2003; Quack et
49 al., 2007; Fuhlbrügge et al., 2016). The open ocean generally shows quite homogeneous, low CHBr_3
50 concentrations, compared to higher concentrations and strong gradients found in coastal and shelf areas
51 (Quack and Wallace, 2003). At the coast, high oceanic concentrations are related to macro algae (Klick
52 and Abrahamsson, 1992) and anthropogenic sources (Boudjellaba et al., 2016) such as power plants
53 (Yang, 2001) and desalination facilities (Agus et al., 2009).

54 Due to sparse measurements and limited process understanding, existing estimates of global air-sea flux
55 distributions of CHBr_3 and other VSLHs are subject to large uncertainties (e.g. Warwick et al., 2006;
56 Palmer and Reason, 2009; Liang et al., 2010; Ordóñez et al., 2012; Stemmler et al., 2013; Ziska et al.,
57 2013; Carpenter et al., 2014). Open ocean background emissions of CHBr_3 are modeled to be around
58 $100 \text{ pmol m}^{-2} \text{ hr}^{-1}$ (Ziska et al., 2013), consistent with simultaneous in-situ measurements of air and water
59 concentrations (Butler et al., 2007; Liu et al., 2013; Fiehn et al., 2017). The spatial and temporal
60 distribution of elevated emissions in coastal and upwelling regions is currently based on very limited
61 observations. Campaigns in these regions suggest that emissions generally increase near coastlines, and
62 that sporadic peak emissions with extremely high values can be found (e.g. Butler et al., 2007; Liu et al.,
63 2013; Fuhlbrügge et al., 2016; Fiehn et al., 2017). Analysis of the measurements suggests that such peak
64 emissions are often of limited spatial extent and cover not more than a distance of 50-100 km along the
65 cruise track. We will use the term ‘elevated emissions’ when describing emissions that are on average
66 up to a factor of 10 larger than the background and ‘hotspot emissions’ for sporadic emissions up to a
67 factor of 100 larger than the background.

68 There are two main approaches to derive the magnitude of VSLH emissions, i.e. “bottom-up” approach
69 (e.g. Quack and Wallace, 2003; Carpenter and Liss, 2000; Butler et al., 2007; Ziska et al., 2013) and
70 “top-down” approach (e.g. Warwick et al., 2006; Liang et al., 2010; Ordóñez et al., 2012). For the
71 “bottom-up” method, measured surface sea water concentrations of VSLHs at the “bottom” (surface) are
72 extrapolated to estimate global emissions. For the “top-down” method, the emissions of VSLHs are
73 constrained by the measured abundances at the “top” (atmosphere) so that model simulations based on



74 the constrained global emission estimates reproduce the observed atmospheric concentrations. These two
75 approaches yield different estimates of the global VSLHs emissions, with the recent “top-down”
76 approaches resulting in generally higher emissions than the recent “bottom-up” approaches.

77 In the tropical ocean waters of the Atlantic, the western Pacific and Indian Ocean, the existence of
78 localized elevated CHBr_3 emissions and hotspots has been confirmed (Butler, et al, 2007; Liu et al., 2013;
79 Krüger and Quack, 2013; Quack and Krüger, 2013; Fiehn et al., 2017). At the same time, these
80 convectively active regions offer an efficient pathway for the vertical transport of short-lived oceanic
81 compounds from the boundary layer to the stratosphere (e.g. Aschmann et al., 2009; Hossaini et al., 2012;
82 Tegtmeier et al., 2012, 2013; Marandino et al., 2013; Liang et al., 2014). Moreover, the Asian monsoon
83 has been recognized as an efficient transport pathway for short-lived pollutants and VSLHs (Randel et
84 al., 2010; Hossaini et al., 2016; Fiehn et al., 2017). Given that elevated oceanic CHBr_3 emissions are
85 expected to occur in the same regions as strong convection, it is of interest to analyze how these elevated
86 emissions impact CHBr_3 in the atmospheric boundary layer, which feeds into the upward transport.

87 Measurements of CHBr_3 abundance in the atmospheric boundary layer show large spatial variability (e.g.,
88 Quack and Wallace, 2003; Montzka and Reimann, 2011; Lennartz et al., 2017). A compilation of
89 available measurements by Ziska et al. (2013) suggests similar CHBr_3 distribution patterns in the
90 atmospheric boundary layer as in the surface ocean, with higher mixing ratios in the equatorial, coastal
91 and upwelling regions. However, given the sparse data base and the uncertainties in the spatial and
92 temporal extent of oceanic emissions, the detailed distribution of boundary layer CHBr_3 , cannot be well
93 constrained (e.g., Hepach et al., 2014; Fuhlbrügge, et al., 2013). On the one hand, the spatial and temporal
94 extent of elevated localized emissions is usually unknown, leading to large uncertainties when estimating
95 their overall magnitudes. On the other hand, the influence of meteorological conditions, distinctive
96 transport patterns and variations of atmospheric sinks, such as the background OH field (e.g. Rex et al.,
97 2014), can be expected to modulate the effect of elevated oceanic sources. Therefore, it is still an open
98 question of the magnitude of elevated and hotspot emissions on the local atmospheric CHBr_3 distribution.
99 Such knowledge is relevant to understand the importance of localized elevated emissions for atmospheric
100 abundances and to interpret existing atmospheric measurements with respect to potential sources and
101 driving factors.

102 In this study, we use observational data from three tropical research cruises, one in the Indian Ocean
103 (OASIS) and two in the western Pacific (TransBrom and SHIVA). We use the Lagrangian particle
104 dispersion model FLEXPART to investigate the transport and atmospheric distribution of VSLHs.
105 Taking bromoform (CHBr_3) as example, we compare the atmospheric signals estimated from the elevated
106 and hotspot emissions measured during the ship campaigns to the distribution derived from only uniform
107 background emissions. The campaigns and the FLEXPART model are introduced in Sect. 2. In Section
108 3, we discuss the distributions and variability of atmospheric CHBr_3 based on uniform background
109 emissions. We present the observed hotspots of CHBr_3 emissions in Section 4.1, and compare the



110 simulated atmospheric mixing ratios resulting from elevated emissions during three campaigns with the
111 background values (Section 4.2). Conclusions are given in Section 5.

112

113 2. Data and Methods

114

115 2.1 Background and in-situ CHBr_3 emissions

116

117 In this study, we distinguish between open ocean background and in-situ CHBr_3 emissions. Open ocean
118 emissions are deduced to be around $100 \text{ pmol h}^{-1} \text{ m}^{-2}$ based on global bottom up scenarios (Quack and
119 Wallace, 2003, Ziska et al., 2013). While emissions for individual regions and seasons can be higher or
120 lower than this, including negative fluxes going from the atmosphere into the ocean, $100 \text{ pmol h}^{-1} \text{ m}^{-2}$
121 represents the typical mean value averaged over all oceanic basins between 60°S and 60°N . The
122 background open ocean emissions exclude by design emissions from coastal, shelf and upwelling regions.

123

124 In-situ oceanic emissions of CHBr_3 have been calculated from the observational data collected during
125 three tropical ship campaigns. The two campaigns TransBrom (October 11th-23rd, 2009, Krüger and
126 Quack, 2013) and SHIVA (November 15th-28th, 2011, Quack and Krüger, 2013) took place in the
127 western Pacific, while the OASIS campaign (July 11th-August 6th, 2014, Fiehn et al., 2017) was
128 conducted in the western Indian Ocean. During each campaign, surface air and water samples were
129 collected simultaneously at regular intervals (every 3 to 6 hours). The emissions were calculated from
130 these co-located data and the instantaneous wind speed (Ziska et al., 2013, Fuhlbrügge et al., 2016, Fiehn
131 et al., 2017). The detailed cruise track and the magnitude of the oceanic CHBr_3 emissions of each
132 campaign is given in Fig. 1. The in-situ emissions include both open-ocean emissions and elevated
133 emissions from coastal, shelf and upwelling regions with our evaluations focusing on the regions of
134 elevated emission.

135

136 2.2 Modeling

137

138 For the simulations of the atmospheric distribution and transport of CHBr_3 , we used the Lagrangian
139 particle dispersion model, FLEXPART (Stohl et al., 2005), which has been validated by previous
140 comparisons with measurements (Stohl et al., 1998; Stohl and Trickl, 1999). Lagrangian particle models
141 such as FLEXPART compute trajectories of a large number of so-called particles, presenting
142 infinitesimally small air parcels, to describe the transport, diffusion and chemical decay of tracers in the
143 atmosphere. The model includes turbulence in the boundary layer and free troposphere (Stohl and
144 Thomson, 1999) and a moist convection scheme (Forster et al., 2007) following the parameterization by
145 Emanuel and Živković-Rothman (1999). The representation of convection in FLEXPART simulations
146 has been validated with tracer experiments and ^{222}Rn measurements (Forster et al., 2007). Chemical or
147 radioactive decay of the transported tracer is accounted for by reducing the particle mass according to a



148 prescribed lifetime of the tracer. Alternatively, the loss processes can be prescribed via OH reaction
149 based on a monthly averaged 3 dimensional OH-field. In this study, we employ FLEXPART version
150 10.0, which is driven by 3-hourly meteorological fields from ECMWF (European Centre for Medium-
151 Range Weather Forecasts) reanalysis product ERA-Interim (Dee et al., 2011) with a horizontal resolution
152 of $1^\circ \times 1^\circ$ and 61 vertical model levels.

153 We performed two kinds of simulations based on the different emission scenarios. The first one used the
154 uniform global background emission, and the second one used in situ emissions observed during
155 individual ship campaigns. Chemical decay of CHBr_3 was simulated by prescribing a lifetime of 17 days
156 during all runs (Montzka and Reimann, 2011). For the background runs, a uniform air-sea flux of 100
157 $\text{pmol h}^{-1} \text{m}^{-2}$ is prescribed over all ocean surface area between 60°S and 60°N . Three runs are conducted
158 covering the time period of the campaigns with a 1-month spin-up period in each case to reach a stable
159 background concentration in the atmosphere.

160 For the in-situ emissions of each campaign, simulations are based on the calculated CHBr_3 air-sea flux
161 (see Fig. 7, detailed description in Sec 4), which is released along the cruise track. The periods of these
162 campaign simulations are the same as the corresponding background simulations with emissions over
163 the whole time period. For each observational data point, an emission grid cell centered on the
164 measurement location is created. These grid cells are designed to be adjacent along the cruise track and,
165 based on the density of the measurements, are about $0.1 - 2.0^\circ$ wide in cruise track direction. The grid
166 cells are chosen to be of a fixed width (0.5° or 1°) in the other direction and thus add up to the narrow
167 band of 0.5° or 1° width centered along the cruise track (Fig. 1). Our design of the emission grid cells
168 assumes that the elevated emissions can extend over a distance of 0.5° - 1° . This choice has been motivated
169 by the spatial variability of the measurements along the cruise track (see also section 4.1 and Fig. 7).
170 Elevated emissions larger than $1000 \text{ pmol h}^{-1} \text{m}^{-2}$ are found at 77 different locations along the three cruise
171 tracks examined in this paper. Out of the 77 measurements, only 11 correspond to singular locations with
172 no adjacent high emissions at the neighboring points. The other 66 measurements cluster together at 18
173 different locations with at least two adjacent observational points showing emissions larger than 1000
174 $\text{pmol h}^{-1} \text{m}^{-2}$. We define the length of such a location of elevated emissions as the distance between the
175 first and last data point with an air-sea flux exceeding $1000 \text{ pmol h}^{-1} \text{m}^{-2}$. Most of the 18 locations extent
176 over a distance larger than 0.5° (13 out of 18) and nearly half are larger than 1° (8 out of 18) supporting
177 our choice of the width of the emissions grid cells. Note that at the same time, the spatial extent of the
178 hotspots is comparable to the wind field resolution that drive our trajectory simulations. The amount of
179 CHBr_3 released from each grid cell is determined by the observational air-sea flux of the corresponding
180 data point and scales with the width of the narrow emission band described above. The specified CHBr_3
181 emission from each cell is kept constant for the duration of the model run and distributed over a fixed
182 number of trajectories. In order to capture the small scale processes (e.g. convection), the large number
183 of 2000, and 20000 trajectories are chosen to be released from each emission grid cell of background run
184 and in-situ run, respectively. Output data in form of CHBr_3 volume mixing ratios available at a user-
185 defined grid, is retrieved at a horizontal resolution of $1^\circ \times 1^\circ$ and $0.5^\circ \times 0.5^\circ$ for background runs and in-



186 situ runs, respectively, at every 100 m from 100 m to 1 km, and every 1 km from 1 km to 20 km every 3
187 hours.

188

189 3. Atmospheric CHBr₃ based on open ocean background emissions

190

191 The atmospheric CHBr₃ mixing ratios diagnosed from the uniform background emissions (referred to as
192 CHBr₃ background mixing ratios hereinafter) vary significantly from campaign to campaign and also
193 within each campaign region. Figures 2 to 4 present several snapshots of the CHBr₃ background mixing
194 ratios and the simultaneous wind fields from ERA-Interim reanalysis for the three campaigns. For
195 TransBrom (Fig. 2), CHBr₃ accumulates south of 15° N with a maximum near the equator, where the
196 wind is weak. In the northern Pacific, which was dominated by an anticyclone centered around 165°E,
197 30°N, the background values are much lower. On the 10th of October 2009, two bands of extremely low
198 wind fields exist, one directly south of the equator and one tilting from 15°N to 5°N, which both coincide
199 with the highest CHBr₃ abundances. On the 20th of October, these two bands collided into one with
200 lowest winds centered around 165°E, where we again find very high values of CHBr₃ of up to 0.8 ppt.
201 For both case studies, highest values are found in the region of the lowest wind speeds or slightly shifted
202 towards the region of strongest wind shear. Regions of high wind speeds, such as the northern Pacific
203 anticyclone, on the other hand do not allow for accumulations and are characterized by very low CHBr₃.
204 For the SHIVA case (Fig. 3), the background CHBr₃ accumulates in a narrow region near Indonesia,
205 with corresponding wind fields smaller than 3 m/s. North of Indonesia, the strong easterly trade winds
206 generally above 10 m/s prevent the accumulation of higher background values within the region. Again,
207 the two case studies illustrate how changes of the wind patterns within a few days drive changes of the
208 background CHBr₃ distribution. Another particular example is the northward extension of the low
209 equatorial winds around 90°E on 16th November 2011, which leads to higher CHBr₃ north of the equator
210 up to 15°N.

211 For OASIS (Fig. 4), the wind speed is higher than in the other two regions and these strong
212 southeast/southwest trade winds associated with the Asian monsoon extend over most of the Indian
213 Ocean. Consistent with the stronger winds, the background values for the OASIS case are significantly
214 lower than for the other two cases, although they also show accumulations in certain regions. These
215 accumulations appear partially in regions of low wind speeds (e.g., near the equator between 70°E and
216 90°E on 17th of July) or in adjacent regions of high wind shear (e.g., north of the equator between 70°E
217 and 90°E for both case studies). For the latter case, the CHBr₃ accumulation also extends into the region
218 of high wind speeds, which is different from the distribution found for the TransBrom and SHIVA
219 regions. This difference occurs because the east coast of the Indian Subcontinent offshore is a region
220 with wind convergence (not shown), which tends to accumulate air masses therein.

221 Given that the accumulation of CHBr₃ background mixing ratios follows in most cases the wind field
222 patterns on a regional scale, we hypothesize that the same relationship holds on a global scale. The global



223 distributions of atmospheric CHBr_3 based on background emissions and wind fields averaged over the
224 time periods of the SHIVA and OASIS cruise are presented in Fig. 5 and 6, respectively. We omit the
225 time period of the TransBrom case, since the background CHBr_3 distribution diagnosed for this period
226 is very similar to background found for the SHIVA period. The global CHBr_3 background mixing ratios
227 (Fig. 5a, and 6a) display a very heterogeneous distribution in spite of the uniform background emission
228 used for the simulations. Accumulations of CHBr_3 are again generally located in the regions of low wind
229 speeds. For the SHIVA period (November 2011), particularly high CHBr_3 background values of 0.3 to
230 0.4 ppt are found along the equator over the Maritime continent, West Pacific, Indian Ocean and at the
231 West coast of Africa, all of which are characterized by particularly low winds. In the Northern and
232 Southeast Pacific, the wind speed is generally higher, and the corresponding CHBr_3 values of less than
233 0.15 ppt are much lower than in the tropical region. For the OASIS period (July/August 2014), the global
234 CHBr_3 distribution is mostly reversed compared to the SHIVA period and high winds over the Indian
235 Ocean and Maritime continent lead to low CHBr_3 abundance in this region. The North Pacific on the
236 other hand, with low wind speeds is now a region of intense accumulation leading to 0.3-0.4 ppt of CHBr_3 .
237 The tropical West Pacific is the only region that experiences relatively low winds during both seasons,
238 and constantly shows high CHBr_3 for the SHIVA and OASIS time periods.

239 The variations of the background CHBr_3 distribution can be generally explained by the seasonal
240 variations of the global wind field. The North Pacific and Northern Indian Ocean are dominated by the
241 East Asia Monsoon and the Monsoon of South Asia, respectively. The East Asia Monsoon is
242 characterized by strong northwesterly flow in boreal winter and weak southeasterly flow in boreal
243 summer due to the reverse of the thermal gradient between land and ocean (Webster, 1987; Ding and
244 Chan, 2005). Therefore, the accumulations of CHBr_3 in the North Pacific occurs during the boreal
245 summer months, rather than during boreal autumn/ early winter (TransBrom time period). The Monsoon
246 of South Asia, on the other hand, is characterized by weak northeasterly winds in boreal winter and
247 strong southwesterly winds in boreal summer (Webster, 1987; Webster et al., 1998). Thus background
248 CHBr_3 accumulation over the Northern Indian Ocean occurs mostly during boreal winter, while during
249 boreal summer (OASIS time period) a low CHBr_3 background can be expected. Because of the light
250 winds of the Inter Tropical Convergence Zone (ITCZ), a belt of relatively high CHBr_3 abundance exists
251 along the equator in the Northern Hemisphere, especially in the tropical Pacific and Atlantic. Strong
252 convection in the ITCZ enhances vertical transport of CHBr_3 out of the boundary layer, but overall the
253 CHBr_3 distribution is dominated by the horizontal wind fields and accompanying transport patterns. Due
254 to the more complex land-sea thermal difference, the seasonal variation of ITCZ in the West Pacific is
255 more significant than in the East Pacific (Waliser and Jiang, 2014). The relatively high accumulations of
256 CHBr_3 in the tropical East Pacific are confined to a narrow region near the equator for both seasons. As
257 for the tropical West Pacific, during boreal winter the ITCZ covers almost the whole Southeast Asia and
258 the high CHBr_3 abundances during SHIVA appear along the east coast of Malaysia. During boreal
259 summer, the ITCZ shifts northward and the high CHBr_3 abundances retreat northwestward.



260 We assume constant CHBr_3 open ocean emissions of $100 \text{ pmol h}^{-1} \text{ m}^{-2}$ for our simulations in order to
261 isolate the impact on the atmospheric CHBr_3 distribution of atmospheric transport patterns versus the
262 impact of varying emission fields. In particular, variations of the wind fields will impact the ocean air-
263 sea flux, and emissions larger than $100 \text{ pmol h}^{-1} \text{ m}^{-2}$ might occur in regions of higher winds with little
264 CHBr_3 accumulation. Such variations can change the background CHBr_3 distribution and may allow for
265 increased mixing ratios in regions of strong winds. In addition to the wind speed, variations in the
266 atmospheric and, more importantly, the oceanic CHBr_3 concentrations can impact the emission strength
267 which can further change the complex atmospheric CHBr_3 distribution.

268

269 **4. Atmospheric CHBr_3 based on hotspot emissions**

270

271 Given the high variability of the atmospheric CHBr_3 background mixing ratios, resulting from
272 atmospheric transport processes (Section 3), it is of interest to analyze if and how much oceanic hotspot
273 emissions might impact this background distribution. In this section, we will use observational data to
274 discuss if oceanic hotspot emissions occur at the same time and location as peak atmospheric mixing
275 ratios or if the two quantities are rather uncorrelated. Furthermore, we will use FLEXPART simulations
276 to compare CHBr_3 mixing ratios that result from background emissions to the increased CHBr_3 mixing
277 ratios that result from localized hotspot emissions.

278

279 **4.1 Observed hotspot emission**

280

281 Oceanic CHBr_3 emissions, atmospheric CHBr_3 mixing ratios and the observed local surface wind speeds
282 are given in Fig. 7 for all three campaigns. The oceanic emissions of CHBr_3 vary substantially from
283 campaign to campaign with mean values of $261 \text{ pmol h}^{-1} \text{ m}^{-2}$ (TransBrom), $1228 \text{ pmol h}^{-1} \text{ m}^{-2}$ (SHIVA),
284 and $912 \text{ pmol h}^{-1} \text{ m}^{-2}$ (OASIS) with standard deviations of $600 \text{ pmol h}^{-1} \text{ m}^{-2}$ (TransBrom), 1460 pmol h^{-1}
285 m^{-2} (SHIVA) and $1159 \text{ pmol h}^{-1} \text{ m}^{-2}$ (OASIS), respectively. All three campaigns show periods with
286 background emissions around $100 \text{ pmol h}^{-1} \text{ m}^{-2}$ and periods with localized elevated and hotspot emissions.
287 For TransBrom, the first two thirds of the campaign show negative (air-to-sea) or very low background
288 CHBr_3 fluxes, while the last third was close to western Pacific islands and is characterized by overall
289 elevated emissions with sporadic hotspots of up to $4000 \text{ pmol h}^{-1} \text{ m}^{-2}$. The SHIVA cruise track, on the
290 other hand, was mostly along the coastline, and low background emissions occur only for brief periods.
291 Most locations showed elevated emissions and hotspots occurred regularly. The OASIS cruise track
292 alternated between open ocean, upwelling and coastal areas, resulting in a large fluctuation between low
293 background and localized elevated emissions. Hotspot emissions during this campaign are largest,
294 reaching values of over $6000 \text{ pmol h}^{-1} \text{ m}^{-2}$.

295 According to the flux parameterization applied here, the air-sea flux is determined mostly by the surface
296 wind speed and the ocean-atmosphere concentration gradient. Highest emissions are expected to occur



297 during periods of high wind speed and large concentration gradients. The wind speed dominates the
298 fluxes for some regions, but not for entire campaigns. During the beginning of the TransBrom campaign
299 (Fig. 7a), the wind speed peaks at over 15m/sec while the corresponding CHBr_3 air-sea flux is low.
300 Higher wind speeds co-occur with high air-sea fluxes at the end of the campaign. For SHIVA (Fig. 7b)
301 and OASIS (Fig. 7c), the relation between wind speed and CHBr_3 emissions is more easily discernable.
302 All three campaigns demonstrate that high fluxes do not always lead to local high CHBr_3 mixing ratios
303 in the surface atmosphere. For example, several hotspots with oceanic emissions over $4000 \text{ pmol m}^{-2} \text{ hr}^{-1}$
304 are found during OASIS, however, corresponding atmospheric mixing ratios are relatively low (~ 2
305 ppt). Vice versa, the highest atmospheric mixing ratios found during OASIS do not coincide with high
306 fluxes, except for the last part of the campaign. These discrepancies suggest that the local atmospheric
307 mixing ratios are driven by a complex interplay of source and loss processes driving the atmospheric
308 mixing ratios of short-lived compounds. A relatively clear connection between elevated oceanic
309 emissions and surface mixing ratios only occurs during the SHIVA campaign and during the last part of
310 the TransBrom campaign (Fig 7a and b).

311 As the campaign data shows a relation between emissions and atmospheric mixing ratios only for some
312 regions, the question arises how much of the atmospheric variability of short-lived compounds such as
313 CHBr_3 is impacted by the emission strengths. In order to quantify the relative contributions of elevated
314 emissions, comparisons between the computed mixing ratios over hotspots and the background
315 abundances in the atmosphere are required. In the subsequent section, we present such comparisons based
316 on the model results.

317

318 **4.2 Comparison of CHBr_3 from background and hotspot emissions**

319

320 In this section, we will compare the concentrations of CHBr_3 due to background and localized elevated
321 emissions as simulated by FLEXPART. First, atmospheric CHBr_3 during all three campaigns is
322 calculated based on the uniform background emission of $100 \text{ pmol h}^{-1} \text{ m}^{-2}$. Second, atmospheric CHBr_3
323 resulting from strong localized emissions is simulated for the three case studies given by the campaigns.
324 Atmospheric CHBr_3 at different altitudes is simulated by FLEXPART, which is driven by the
325 meteorological data from ECMWF. The signatures of dynamical processes such as wind regimes,
326 weather phenomena (e.g., typhoons) and convection are captured by the model simulation and can be
327 detected in the CHBr_3 distribution (Fig. 8). For example, during the TransBrom campaign, the cruise
328 encountered several tropical storms in the western Pacific, one of which (Lupit, around October 14th,
329 2009) developed into a super typhoon within several days (Krüger and Quack, 2013). As shown in Fig.
330 8, an elevated CHBr_3 accumulation representing the structure of typhoon Lupit is clearly visible in the
331 background distribution of CHBr_3 at 500 m (Fig. 8d) altitude in the northern part of the western Pacific.
332 This structure is still clear at 5 km altitude (Fig. 8a), although with a weaker magnitude. For CHBr_3
333 emitted from localized elevated sources (Figures 8b, c, e, and f), such large scale structures are not



334 discernible due to the small spatial extent of the 0.5° or 1° emission cells and thus the limited amount of
335 overall released CHBr_3 . A clear blob of higher abundances of atmospheric CHBr_3 can be seen in the
336 southern part of the western Pacific near Indonesia resulting from one of the hotspot emissions observed
337 during TransBrom (Fig. 1b). However, the background CHBr_3 in this area is also high in this low-wind
338 area, and thus the atmospheric signal of the up to 20 times stronger hotspot emissions (Fig. 7a) is
339 detectable for neither the 0.5° nor the 1° wide emission cells when compared to the background. Note
340 that the modelled atmospheric mixing ratios from both sources, hotspot and background emissions, are
341 smaller than the mixing ratios observed along the cruise track (Figure 7) suggesting stronger nearby
342 emissions not covered in our scenarios and observations. The signature of the hotspot emissions remains
343 in the boundary layer and cannot be seen at 5 km altitude.

344 Fig. 9 shows the atmospheric CHBr_3 mixing ratios during the SHIVA campaign. For the SHIVA case,
345 the distribution patterns of the background CHBr_3 abundances at 500 m altitude also show a very strong
346 spatial variability, despite the uniform emissions. Highest CHBr_3 background mixing ratios around 120°
347 E near the equator of up to 0.5 ppt are smaller than background values found during TransBrom of up to
348 0.8 ppt. The atmospheric signal of the localized elevated emissions is much stronger than during
349 TransBrom due to stronger emissions on the one hand and smaller background mixing ratios on the other
350 hand. First, for the 0.5° wide emission grids, two highly localized, atmospheric CHBr_3 peaks appear
351 close to the coast line near the equator around 105° E with a maximum value around 0.4 ppt. These
352 signals occur in a spot where the background is very low (0.2 ppt). However, at the same time they are
353 smaller than the maximum background values of up to 0.5 ppt in nearby regions (Fig. 9d). If the width
354 of the emission grids is extended to 1° , the localized CHBr_3 peaks mentioned above grow into two distinct
355 blobs near the equator of up to 0.8 ppt. These maxima along the first half of the cruise track are apparently
356 larger than the regional background concentrations (Fig. 9f). Elevated emissions during the second half
357 of the campaign with several hotspot events, on the other hand, do not show such clear atmospheric
358 signals right above.

359 At 5 km altitude, the overall background values are slightly lower, but the maximum mixing ratios are
360 still comparable to altitudes below due to the intense convection in the general region of the SHIVA
361 campaign (Fuhlbrügge et al., 2016). For the regions of localized elevated emissions, the convection is
362 less effective and maximum mixing ratios at 5 km are about 50% smaller compared to the values in the
363 boundary layer. Krystofiak et al. (2018) calculated the fractions of convective-contributed trace gases
364 from boundary layer to the upper troposphere using airborne measurements during the SHIVA campaign
365 and reported an even smaller fraction of boundary layer CHBr_3 in the upper troposphere (about 15% due
366 to convection). Thus only the signal of the 1° wide emission cells can be detected at 5 km, while assuming
367 that the emissions cover a smaller region of 0.5° width will render their impact in the free troposphere
368 negligible.

369 Due to the dominant southwest monsoon over the Northern Indian Ocean in boreal summer, the resulting
370 atmospheric abundances of the OASIS case (Fig. 10) for both scenarios, background and localized



371 emissions, are much lower than for the other two campaigns. This is particularly surprising for the OASIS
372 hotspot emissions, which are in many cases larger than hotspot emissions during TransBrom or SHIVA.
373 In the open ocean, the atmospheric enhanced CHBr_3 mixing ratios resulting from the 0.5° (1°) wide
374 localized emission runs reach only 0.1 (0.2) ppt in a narrow belt near 60°E and are mostly smaller than
375 the background (around 0.15 ppt). An exception occurs near the coast of Madagascar, where both
376 background and hotspot emissions accumulate in the atmosphere. Maximum background values reach
377 up to 0.25 ppt and the hotspot signals peak with values of 0.3 ppt (0.5° wide emission cells) to 0.6 ppt
378 (1° wide emissions cells). These clear atmospheric signals of hotspot emissions are driven by the
379 enhanced coastal emissions near Madagascar. At 5 km altitude, atmospheric background values are very
380 low, and the hotspot contributions are close to zero.

381 In summary, the observed emissions during the three cruises were significantly higher than the
382 background of $100 \text{ pmol m}^{-2} \text{ hr}^{-1}$. Our results show that such strong oceanic sources are not necessarily
383 detectable in the atmosphere, where transport processes can sometimes mask the impact of oceanic
384 emissions on the atmospheric CHBr_3 distribution.

385

386 5. Summary and Discussion

387

388 In this study, we simulated atmospheric CHBr_3 abundances that result from uniform marine background
389 emissions compared to hotspot emissions using the Lagrangian dispersion model FLEXPART.

390 The simulations demonstrate that uniform background emissions from the ocean result in a highly
391 variable atmospheric CHBr_3 distribution with accumulations taking place in regions of low wind speed.
392 This relation holds on regional and global scales revealing atmospheric transport processes as important
393 drivers of the distribution of short-lived trace gases with lifetimes in the range of days to weeks. The
394 relation between atmospheric background and wind patterns described here will allow us to better predict
395 the seasonal and regional characteristics of the tropospheric CHBr_3 distribution. Such knowledge will
396 provide valuable information for analyzing and interpreting atmospheric data from ship and aircraft
397 campaigns. For example, our results illustrate that low atmospheric CHBr_3 abundances cannot
398 necessarily be used to draw conclusions about the oceanic source strength below.

399 Comparisons between atmospheric CHBr_3 resulting from background and peak emissions suggest that
400 the impact of localized elevated emission on the atmospheric CHBr_3 distribution depends on their relative
401 strength, on their location and on the time of emission. The “visibility” of elevated emissions in the
402 atmospheric CHBr_3 distribution varies significantly between three cruises in the West Pacific and Indian
403 Ocean. In the open ocean, signals of elevated emissions can hardly be distinguished from the background
404 CHBr_3 distribution even for elevated sources extending over 1° wide source regions along the cruise
405 tracks. Near the coast, however, signals of elevated emissions are often strong enough to be distinguished
406 from the background. In particular, some of the hotspot emissions up to 100 times larger than the
407 background can be detected in the atmosphere. However, individual cases show that it is not necessarily



408 the largest hotspot that gives a clear signal, but that the tug of war between fast advective transport and
409 local accumulation at the time of emission is also important.
410 The constant background emissions of $100 \text{ pmol m}^{-2} \text{ hr}^{-1}$ used in our study are based on a simplified
411 scenario and do not take coastal and upwelling maxima into account. Realistic oceanic emissions are
412 much more complex with large gradients in the above mentioned regions. Nevertheless, our results
413 demonstrate that atmospheric CHBr_3 signals, produced by localized elevated and even hotspot emissions,
414 orders of magnitudes larger than the average open ocean emissions, can be obliterated by the highly
415 variable atmospheric background. That is to say that transport variations of the atmosphere itself are
416 sufficient to produce high concentrations in certain regions and that high concentrations of VSLH in the
417 atmosphere do not always guarantee a strong local or regional source. For observational and modelling
418 studies of VSLH and other short-lived compounds, the impact of atmospheric transport patterns that are
419 identified here can be used for the interpretation of trace gas distributions and variability.



420 **Data availability**

421 The emission data of cruise campaigns are available at Pangaea (<http://www.pangaea.de>). FLEXPART
422 output can be inquired from the authors.

423

424 **Author contribution**

425 Y. Jia, S. Tegtmeier designed the model experiments. Y. Jia carried out the FLEXPART calculations and
426 produced the figures. Y. Jia and S. Tegtmeier wrote the manuscript with contributions from all co-authors.

427

428 **Competing interests**

429 The authors declare that they have no conflict of interest.

430

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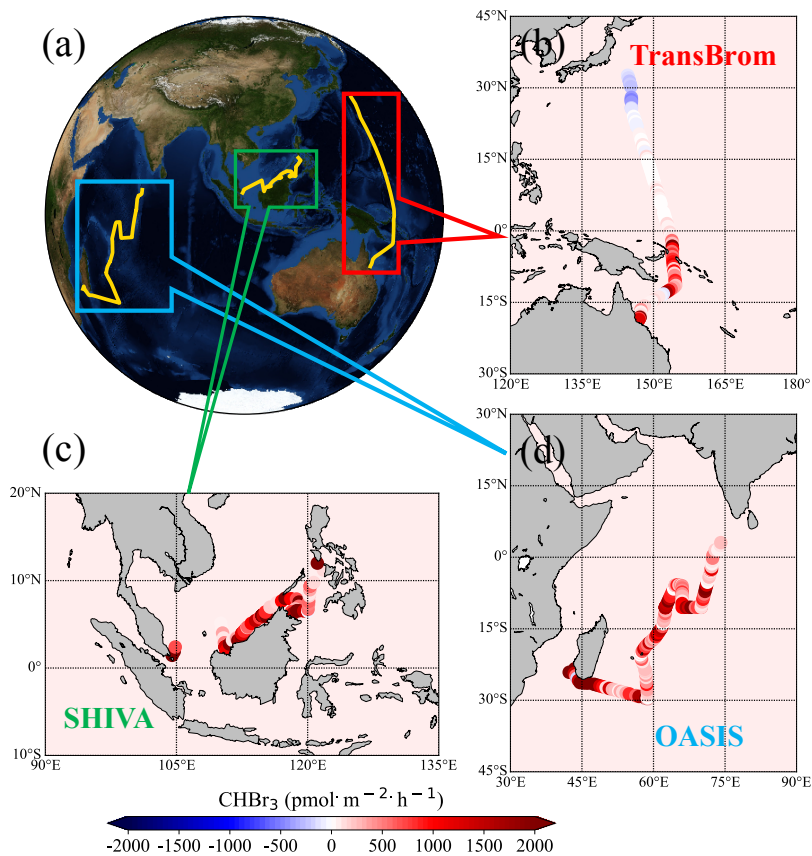
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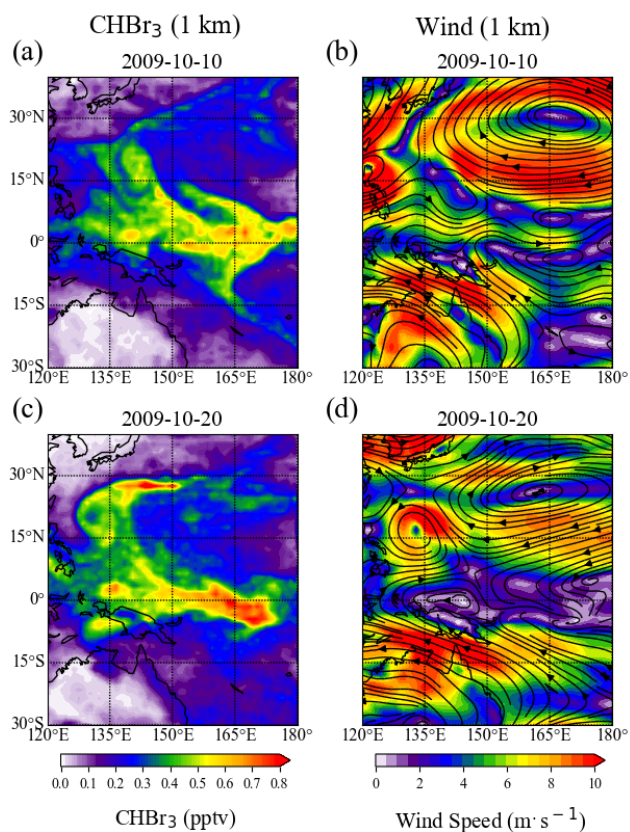
630 **Fig 1.** Cruise tracks of the three campaigns in the Indian Ocean and Western Pacific (a)
631 and CHBr_3 emissions (b, c, d) used in the model simulation. Global background
632 emissions ($100 \text{ pmol m}^{-2} \text{ hr}^{-1}$) and observed emissions along the tracks of the three
633 research cruises TransBrom (b), SHIVA (c), and OASIS (d).

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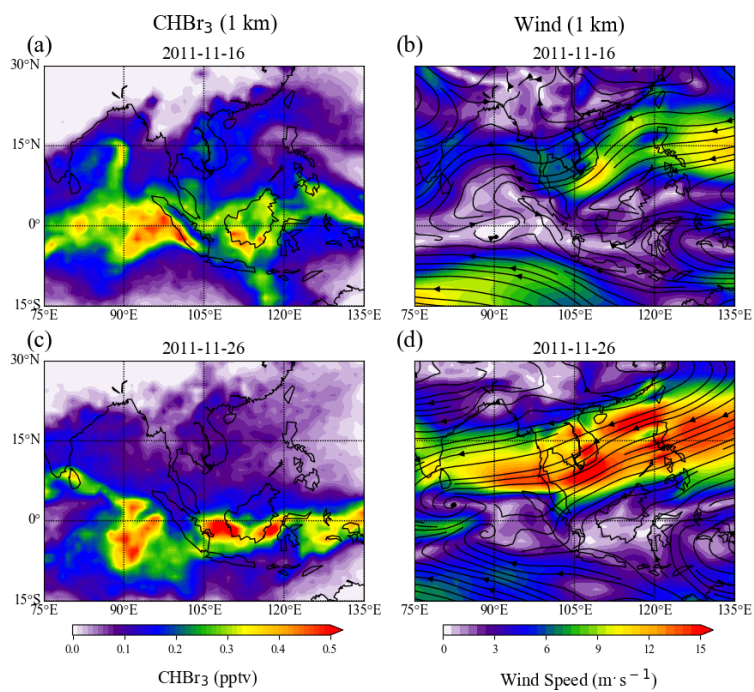
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640 **Fig. 2.** Two snapshots of spatial distributions of atmospheric CHBr₃, derived from
641 uniform oceanic background emissions of $100 \text{ pmol m}^{-2} \text{ hr}^{-1}$ (a, c), and ERA-Interim
642 reanalysis wind fields (b, d) at 1 km altitude during TransBrom. The wind speed is
643 denoted by color shades and the directions are denoted by the stream lines.

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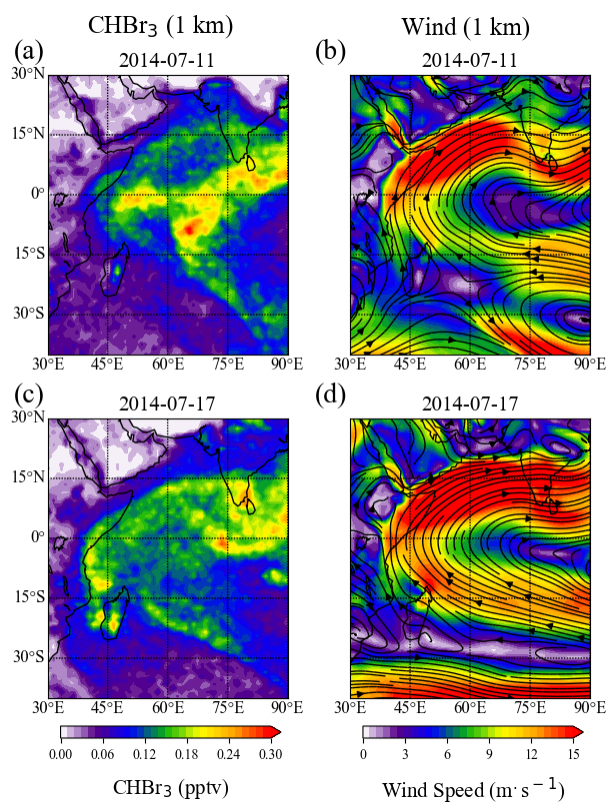


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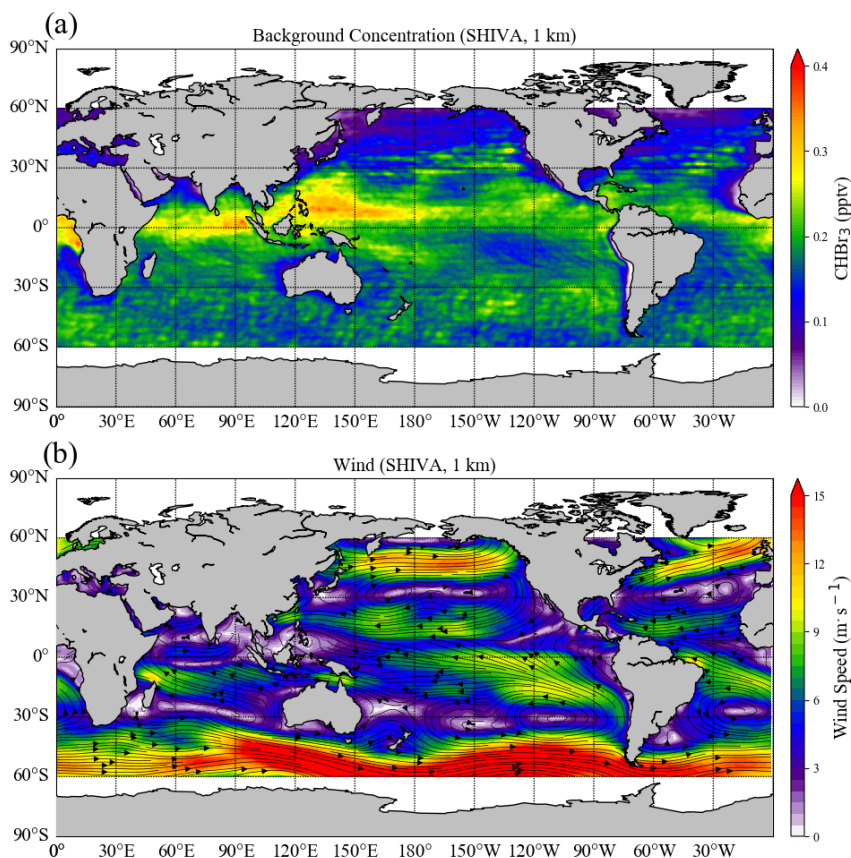
647 **Fig. 3.** Same as Fig. 2, but for SHIVA case.

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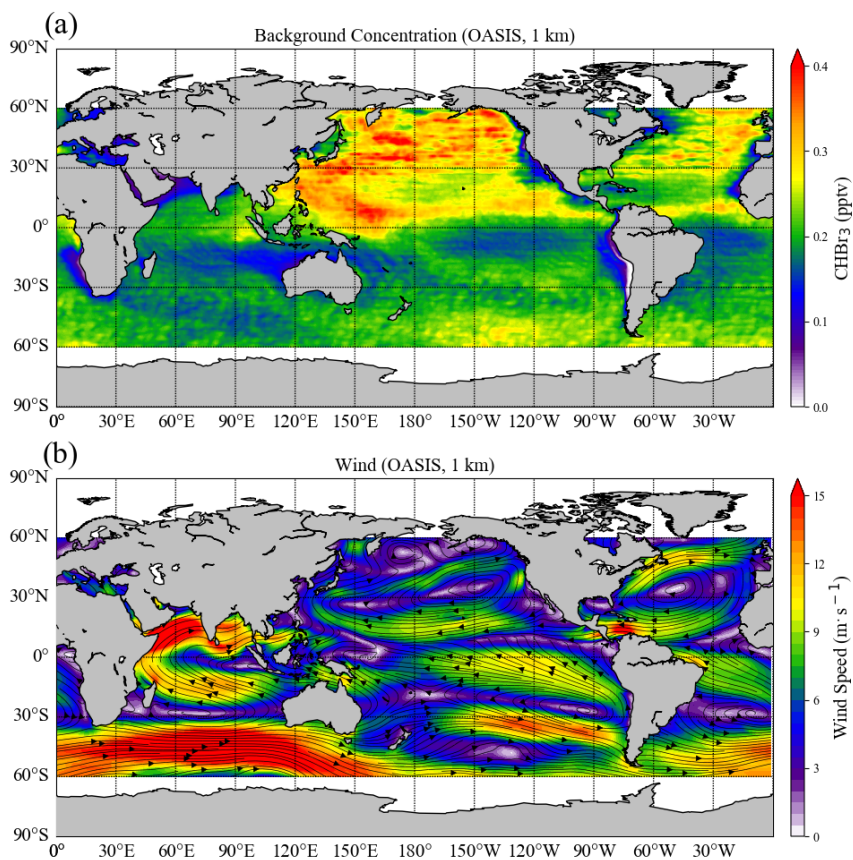
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651 **Fig. 4.** Same as Fig. 2, but for OASIS case.
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654 **Fig. 5.** Global distributions of CHBr_3 mixing ratios based on oceanic background
655 emissions (a), and ERA-Interim reanalysis wind fields (b) averaged during the time
656 period of the SHIVA cruise at 1 km. The wind speeds are denoted by color shades and
657 the directions are denoted by the stream lines.



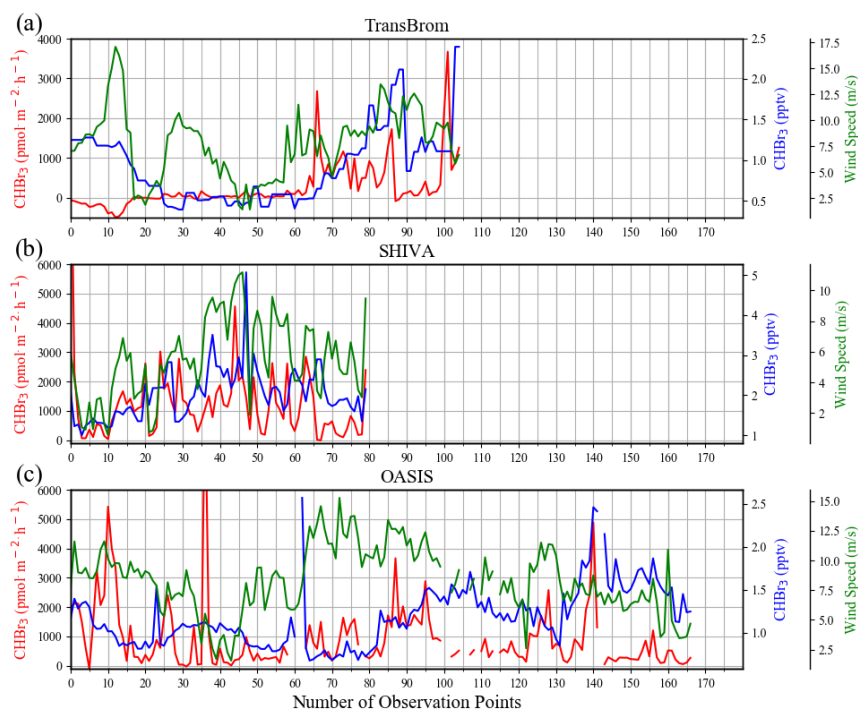
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659 **Fig. 6.** Same as Fig. 5 but for OASIS case.

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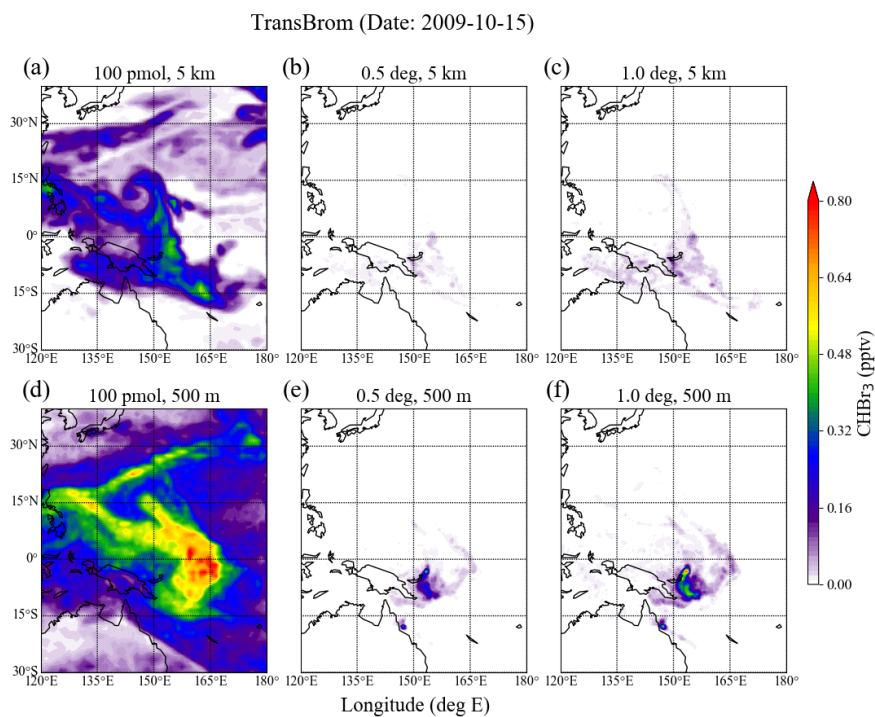


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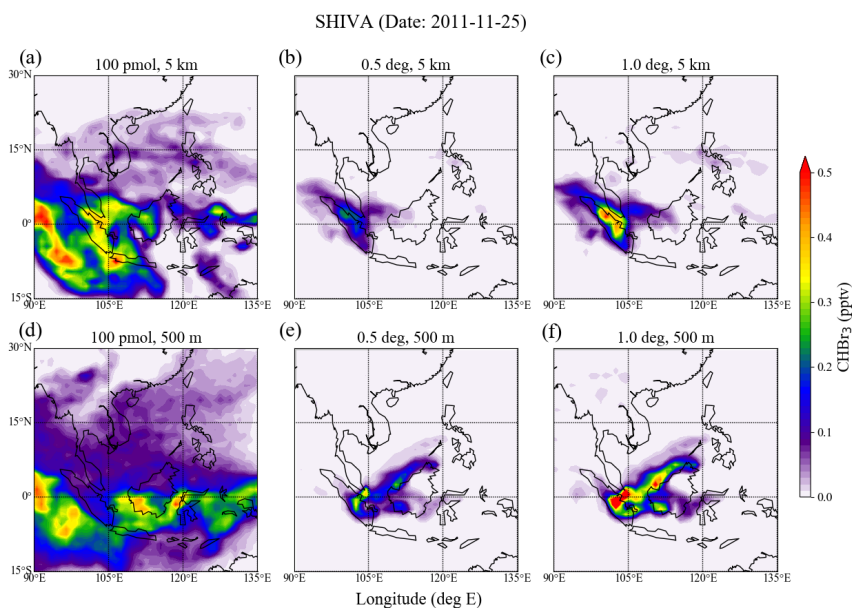
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664 **Fig. 7.** Surface wind speeds (green), CHBr_3 air-sea flux (red), and atmospheric mixing
665 ratios of CHBr_3 near surface (blue) observed during TransBrom, SHIVA, and OASIS
666 campaigns.

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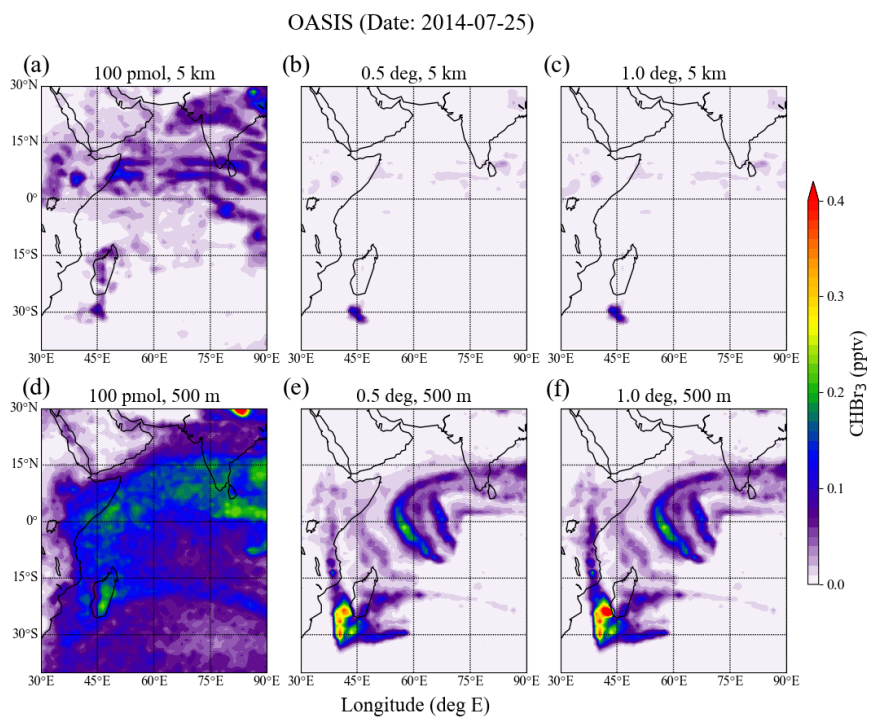


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669 **Fig. 8.** Atmospheric CHBr₃ mixing ratios at different altitudes (500 m and 5 km)
670 simulated for the time period of the TransBrom campaign. Simulations are based on
671 background emissions (a, d), and elevated emissions observed during the campaign for
672 0.5° (b, e) and 1° (c, f) wide emission grid cells.



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Fig. 9. Same as Fig. 8, but for the SHIVA campaign.



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677 **Fig. 10.** Same as Fig. 8, but for the OASIS campaign.
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