

We have made the change to the discussion about the impact of the extratropical emissions. The paragraph now reads as follows:

One aim of this study is to explore the effect of model resolution on the transport of CHBr_3 into the TTL following emissions in tropics. Accordingly from now on, we use the Uniform CHBr_3 tracers (Figure 1 (a) and (c)), rather than Uniform-50, at the two different model resolutions. Recall that, at either resolution, the Uniform tracer has 70% of its emissions in the tropics. By using separate tracers for tropical and extratropical emissions, the contribution of extratropical emissions to the total TTL mixing ratio is found to be small (~10% for the Uniform tracer in the HR model). The effect of extra-tropical emissions (defined here as 20° - 50° N/S) on the CHBr_3 TTL mixing ratios can, to first order, be discounted.

We have not added the information about the boundary layer concentrations. We propose adding that when the first author returns to work.

1 **On the emissions and transport of bromoform: sensitivity to model**
2 **resolution and emission location**

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8 **Abstract**

9 Bromoform (CHBr₃) is a short-lived species with an important but poorly quantified ocean
10 source. It can be transported to the Tropical Tropopause Layer (TTL), in part by rapid,
11 deep convective lifting, from where it can influence the global stratospheric ozone budget.
12 In a modelling study, we investigate the importance of the regional distribution of the
13 emissions and of model resolution for the transport of bromoform to the TTL. We use two
14 idealised CHBr₃ emission fields (one coastal, one uniformly distributed across the oceans)
15 implemented in high and coarse resolution (HR and CR) versions of the same global
16 model and focus on February as the period of peak convection in the West Pacific. Using
17 outgoing long-wave radiation and precipitation as metrics, the HR version of the model is
18 found to represent convection better. In the more realistic HR model version, the coastal
19 emission scenario leads to 15-20% more CHBr₃ in the global TTL, and up to three times
20 more CHBr₃ in the TTL over the Maritime Continent, than when uniform emissions of the
21 same tropical magnitude are employed. Using the uniform emission scenario in both
22 model versions, the distribution of CHBr₃ at 15.7 km (approximately the level of zero net
23 radiative heating) is qualitatively consistent with the differing geographic distributions of
24 convection. However, averaged over the whole tropics, the amount of CHBr₃ in the TTL in
25 the two model versions is similar. Using the coastal scenario, in which emissions are
26 particularly high in the Maritime Continent because of its long coastlines, the mixing ratio
27 of CHBr₃ in the TTL is enhanced over the Maritime Continent in both model versions. The
28 enhancement is larger, and the peak in CHBr₃ mixing ratio occurs at a higher altitude, in
29 the HR model version. Our regional-scale results indicate that using aircraft measurements
30 and coarse global models to infer CHBr₃ emissions will be very difficult, particularly if (as is
31 possible) emissions are distributed heterogeneously and in regions of strong convective
32 activity. In contrast, the global-scale agreement between our CR and HR calculations
33 suggests model resolution is less vital for studies focussed on the transport of bromine into
34 the global stratosphere.

35 **1. Introduction**

36 Very short-lived halogenated substances (VSLS) are thought to make a significant but
37 uncertain contribution to bromine in the stratosphere (5 ± 3 ppt [i.e. ~10-40%] Br,
38 Carpenter, Reimann et al., 2014). Much of this uncertainty is linked to the contribution of
39 bromoform (CHBr₃), which has both the shortest lifetime and the largest emissions of the
40 commonly observed brominated VSLS.

41 The short lifetime of CHBr_3 (~15 days in the tropical boundary layer; Carpenter, Reimann
42 et al., 2014) means that measurements in a particular location can only be used to
43 constrain emissions over relatively small areas of the globe (e.g., Ashfold et al., 2014), and
44 inventories are therefore uncertain (Quack and Wallace, 2003). To illustrate, recent
45 estimates of total global emissions, constructed using various methodologies, range
46 between 120-200 Gg Br yr^{-1} (Ziska et al., 2013) and ~800 Gg Br yr^{-1} (Yokouchi et al.,
47 2005; Butler et al., 2007; O'Brien et al., 2009). Emissions from the oceans are believed to
48 be the major source, but the relative importance of coastal and open ocean emissions is
49 unclear, with uncertainty here linked to the lack of information on the distribution and
50 relative strength of the dominant macro- and micro-algal sources (Ordonez et al., 2012;
51 Stemmler et al., 2015).

52 Model estimates of the contribution that CHBr_3 makes to Br in the stratosphere vary due to
53 the assumed emission distribution (Hossaini et al., 2013), and also due to the treatment of
54 chemical transformations of so-called product gases (e.g., Aschmann and Sinnhuber,
55 2013). A further source of uncertainty of Br injection into the stratosphere, again important
56 because of the short CHBr_3 lifetime, is its sensitivity to model representations of
57 convective transport (e.g., Liang et al., 2014). This is likely to be particularly important
58 when emissions and convective transport are spatially heterogeneous, and possibly co-
59 located (e.g., Tegtmeier et al., 2012).

60 The peak outflow of convection occurs at altitudes around 12-13 km (corresponding to a
61 potential temperature of ~340 K and a pressure of ~200hPa), and the tropical tropopause
62 layer (TTL) is the transition zone between these altitudes and the stratified stratosphere at
63 ~18 km (~380 K, ~90 hPa) (Levine et al., 2007; Randel and Jensen, 2013). During
64 northern hemisphere winter the strongest convection is found over the West Pacific and
65 Maritime Continent, above the warm waters of the Tropical Warm Pool (Gettelman et al.,
66 2002). This is thought to be the region of strongest transport of short-lived species from
67 the ocean surface to the TTL (e.g. Levine et al., 2007).

68 Inherent to uncertainty around modelled convective transport is the spatial resolution of a
69 model. The horizontal distribution of convection is less realistic in models with a coarse
70 resolution (e.g., Russo et al., 2011; Chemel et al. 2015). In particular, coarse resolution
71 models fail to correctly represent geographical features around the Maritime Continent,
72 where CHBr_3 emissions might be large (Pyle et al., 2011), such as coastlines (Schiemann
73 et al., 2014) and orography (Kirshbaum and Smith, 2009). As a consequence, they fail to
74 resolve small-scale dynamical features such as sea breezes which often drive local
75 circulation and convective development in coastal areas (Qian, 2008). In contrast, the
76 vertical extent of convection and the associated vertical transport appears to depend more
77 strongly on the convection parameterisation rather than the model horizontal resolution
78 (e.g. Hoyle et al., 2011).

79 Thus far, global model studies related to CHBr_3 emissions and convective transport have
80 generally employed coarse ($> 2^\circ$) horizontal grids, the resolution used in most climate
81 model studies. Given the sensitivity of convection to resolution outlined above, can we
82 trust these low resolution models when they are used to construct 'top-down' emission
83 inventories (e.g. Warwick et al., 2006)? Are low resolution models suitable for evaluation of

84 the accuracy of those inventories against observations (e.g., Hossaini et al., 2013), or for
85 simulating transport of CHBr_3 towards the stratosphere in convectively active regions?

86 To begin to address these issues, in this study we employ a conventional, coarse
87 resolution version and a high resolution version of the same global model to address two
88 main questions: (i) to what extent does transport of CHBr_3 to the TTL depend on model
89 resolution, and ii) how does the efficiency of transport of CHBr_3 in the two model versions
90 differ when the tropical oceanic emissions are either spatially heterogeneous, being
91 concentrated along shallow coastlines or are uniform across all oceans?

92 In section 2 (Methodology), the model set-up is described. The idealised CHBr_3 emission
93 scenarios used are then discussed in Section 3, and the results regarding the quality of the
94 model convection and its effect on CHBr_3 transport are given in Section 4. Finally in
95 Section 5, we summarise the main findings and discuss the implications, particularly for
96 current estimates of global CHBr_3 emissions.

97 **2. Methodology**

98 Model integrations are performed using the UK Chemistry and Aerosols (UKCA) model
99 (Telford et al., 2010; Archibald et al., 2011; O'Connor et al., 2014), coupled to an
100 atmosphere-only version of the UK Met Office Unified Model (UM version 7.3) (Hewitt et
101 al., 2011).

102 A tropospheric chemistry scheme, described in Telford et al. (2010) and O'Connor et al.
103 (2014), is used to represent chemical cycles of Ox, HOx and NOx as well as the oxidation
104 of CO and other non-methane hydrocarbons as previously described in Zeng and Pyle
105 (2003). The oxidation of isoprene is included by implementation of the condensed Mainz
106 Isoprene Mechanism (MIM) as described in Pöschl et al. (2000). Photolysis rates for
107 photochemical reactions are calculated using the fast-JX photolysis scheme (Neu et al.,
108 2007; Telford et al., 2013). For this study, a bromoform tracer has been added to the
109 existing chemistry scheme. Its oxidation is determined by photolysis (Sander et al., 2006)
110 and reaction with the model-calculated OH (De More et al., 1997). After oxidation, the
111 bromine atoms are ignored, playing no further part in the model chemistry.

112 Present day surface emissions for the chemical species are generated from the emission
113 data set of Lamarque et al. (2010), as developed for the IPCC fifth assessment report.
114 Isoprene emissions are taken from the POET database (Granier et al., 2005; Olivier et al.,
115 2003). The general circulation for the periods under analysis is forced by prescribing
116 monthly mean sea surface temperatures and sea ice cover from the AMIP data set
117 (<http://www-pcmdi.llnl.gov/projects/amip>).

118 The model is run in two different configurations. Firstly, coarse resolution (CR) integrations
119 are performed at a horizontal resolution of 3.75° in longitude \times 2.5° in latitude (gridbox size
120 ~ 300 km) with 60 sigma-height hybrid levels (~ 80 km top). These levels follow earth's
121 surface in the lower troposphere and transition to constant pressure surfaces in the
122 stratosphere and above. Secondly, high resolution (HR) integrations are carried out at a
123 horizontal resolution of $0.56^\circ \times 0.375^\circ$ (gridbox size ~ 40 km) with 63 sigma-height hybrid
124 levels (~ 40 km top). Horizontally, the area of an HR gridbox is ~ 40 times smaller than a

125 CR gridbox, while vertically the average resolution in the troposphere is 600 m for CR
 126 compared to 360 m for HR. Note that the two model configurations are typically optimised
 127 for climate and weather forecast integrations respectively; therefore the impact of these
 128 further differences, which is difficult to disentangle from the simple effect of grid resolution,
 129 will also be reflected in our analysis.

130 One CR integration is performed spanning 10 years (1996-2005) following a 1-year spinup
 131 period while there are five separate HR timeslice integrations for the month of February
 132 and the years 1996, 1998, 2000, 2002 and 2005. A 4 month HR spin-up run is performed
 133 to initialise chemical fields (including bromoform tracers) prior to the February runs. A
 134 summary of the model integrations can be found in Table 1. Each integration is run with
 135 both a uniform ocean emission distribution and a coastal emission distribution, as
 136 discussed in the next section.

137 Table 1 – Characteristics of models and simulations.

Name	Horizontal	Vertical	Run period	Tracers	Tropical emissions (Gg/yr)
CR	3.75°x2.5°	60 levels (80 km top)	1996-2005	Uniform	280
				Coastal	200
HR	0.56°x0.375°	63 levels (40 km top)	Feb '96, '98, '00, '02, '05	Uniform	280
				Coastal	200
				Uniform_50	200

138

139 3. Bromoform emissions

140 In order to address the sensitivity of bromoform transport to model resolution, we designed
 141 two idealised bromoform emission datasets, both with a total of 400 Gg/yr of bromoform
 142 emitted. The two idealised emission datasets are prescribed as follows:

- 143 a) *Uniform* – emissions are uniformly distributed in model ocean gridboxes with 70% of
 144 the emissions in the tropics and the rest in the extra-tropics, in accordance with
 145 Scenario 3 in Warwick et al. (2006). For the purpose of this paper we define the
 146 tropics as the region between 20°S-20°N and the extra-tropics as the region
 147 between 20-50°N/S.
- 148 b) *Coastal* – the Smith and Sandwell Global Seafloor Topography (Smith and
 149 Sandwell, 1997) was used to identify shallow sea areas (defined as having a depth
 150 less or equal to 200m) on the HR model grid. Emissions were distributed equally in
 151 all shallow-sea gridboxes between 50°S and 50°N, which resulted in 50% of
 152 emissions in the tropics and 50% in the extra-tropics. These emissions were then
 153 interpolated on the CR grid with an area-averaging technique, ensuring that the
 154 total amount emitted in similar domains (i.e. in the tropics and extra-tropics) is the
 155 same at both resolutions. Coastal emissions in the Maritime Continent are 29% of
 156 the total coastal emissions and 57% of the tropical coastal emissions.

157 The distribution of coastal and uniform emissions at both model resolutions is presented in
 158 Figure 1. A third tracer was also used for comparative purposes:

159 c) since the amount of bromoform emitted in the tropics is different in the coastal and
160 uniform tracers, an additional uniform bromoform tracer was used in HR runs
161 (*Uniform_50*), with only 50% of emissions distributed in the tropics. This allows us
162 to compare directly the coastal and uniform_50 concentrations in the Tropics and
163 therefore investigate the sensitivity of model convective transport on the spatial
164 location of emissions.

165 **4. Results**

166 **4.1 Convection characteristics**

167 Monthly mean maps of Tropical Rainfall Measuring Mission (TRMM) observations of
168 precipitation and from the Atmospheric InfraRed Sounder (AIRS) observations of out-going
169 long-wave radiation (OLR) are shown in Figure 2 for February 2005 chosen as a
170 representative February (see below) for which an HR run was available. Equivalent
171 quantities for the coarse and high resolution model runs are also shown. The most obvious
172 differences compared to the observations are with the CR integration and are: (a) the
173 misplaced location of the convection in the CR run in the West Pacific with the maximum
174 being incorrectly restricted to a narrow band associated with the Inter-Tropical
175 Convergence Zone (ITCZ); and (b) the overly strong continental convection in the CR
176 model over S. America and S. Africa. This can be explained by differences in low level
177 circulation and surface moisture fluxes between CR and HR. With its larger grid box size,
178 CR integrations fail to properly represent the sharp gradients between land and sea
179 around the islands in the Maritime Continent and this leads to a poor representation of
180 wind convergence and sea breezes. Over large continents, precipitation is often
181 overestimated in CR, which leads to a positive feedback cycle of moister surface and
182 further enhanced convection. Using either precipitation or OLR as a measure of model
183 performance, it is evident that the high resolution integrations perform better.

184 In the rest of this study, we concentrate on February 2005 to study differences in TTL
185 bromoform concentrations arising from the different model resolutions or different
186 distributions of emissions. OLR was anomalously low over much of the Maritime Continent
187 and northern Australia in February 2005, which was in the declining phase of a weak
188 ENSO event (Oceanic Nino Index of 0.4 –
189 http://www.cpc.ncep.noaa.gov/products/analysis_monitoring/ensostuff/ensoyears.shtml).
190 Nevertheless, a comparison of the 5 year average of the different high resolution runs and
191 the 10 year average from the coarse resolution run shows broadly similar results (not
192 shown), so the choice of year is not crucial. A comparison with the multi-year averages is
193 given at the end of Section 4.2.2 to illustrate the relative magnitude of the variability
194 calculated with the different model resolutions and emission scenarios.

195 **4.2 Bromoform transport**

196 In this section we first use the more realistic HR model to compare the cases with coastal
197 emissions and uniform emissions. We then look at the effect of the model resolution.

198 **4.2.1 Coastal versus uniform emissions at high resolution**

199 Figure 3 shows the mixing ratio of CHBr_3 , comparing the Uniform_50 and Coastal tracers,
200 at 15.7 km altitude in February 2005 calculated using the high resolution model. An
201 altitude of 15.7 km is used as it is the model level close to, but above, the level of zero net
202 radiative heating above which air will ascend into the stratosphere (see Russo et al. (2011)
203 for a detailed discussion about the height of the zero radiative heating level). Figure 3(a)
204 shows the case where the emissions of CHBr_3 are uniformly distributed across the tropical
205 oceans, while Figure 3(b) shows the case for an equal amount of emissions concentrated
206 in the shallow coastal regions. The mixing ratios in the TTL around the Maritime Continent
207 are noticeably greater for the coastal emissions. This can be seen more clearly in the
208 panels (c) and (d) where the average vertical profiles are shown for the Tropics and for the
209 Maritime Continent (indicated by the rectangle in the top two plots). The peak values for
210 the Coastal tracer at ~15 km are over twice as large as for the Uniform-50 tracer over the
211 Maritime Continent. This is consistent with the enhanced emissions in this region due to
212 the long coastlines (and hence large area with low ocean depth) associated with the
213 islands (see Fig 1) combined with the enhanced upward transport in convection over this
214 region. Globally, the CHBr_3 mixing ratio in the TTL is approximately 15-20% higher when
215 the coastal emissions are used. This is again due to a shorter time between emission and
216 lofting into the TTL when the coastal emissions are collocated with the convection, leading
217 to less chemical degradation of CHBr_3 in the low and middle troposphere. These results,
218 for February 2005, are robust across the 5 different years studies in the HR integrations.

219 The large local differences between CHBr_3 calculated in the TTL for the different model
220 emission distributions (but with the same magnitude of emissions across the tropics) has
221 important implications for emissions derived from aircraft measurements. We discuss this
222 further in Section 5.

223 4.2.2 High resolution versus coarse resolution.

224 One aim of this study is to explore the effect of model resolution on the transport of CHBr_3
225 into the TTL following emissions in tropics. Accordingly from now on, we use the Uniform
226 CHBr_3 tracers (Figure 1 (a) and (c)), rather than Uniform-50, at the two different model
227 resolutions. Recall that, at either resolution, the Uniform tracer has 70% of its emissions in
228 the tropics. By using separate tracers for tropical and extratropical emissions, the
229 contribution of extratropical emissions to the total TTL mixing ratio is found to be small
230 (~10% for the Uniform tracer in the HR model). The effect of extra-tropical emissions
231 (defined here as 20°-50°N/S) on the CHBr_3 TTL mixing ratios can, to first order, be
232 discounted.

233 Figure 4 (a) and (b) show the 15.7 km CHBr_3 field for February 2005 modelled with
234 Uniform emissions from the high and coarse resolution model runs, respectively. The
235 overall mixing ratio patterns are consistent with the different distributions in convection
236 over the oceanic source regions for CHBr_3 . There is a sharp peak in bromoform mixing
237 ratios along the ITCZ in the CR calculation, associated with the unrealistic peak in
238 convection discussed in Section 4.1. When averaged over the whole of the Tropics, the
239 vertical profiles shown in the panels (c) and (d) show little difference. However, we do find
240 ~10-15% higher mixing ratios at 15.7 km in the coarse resolution run over the Maritime
241 Continent.

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245 Larger differences are seen between the coarse and high resolution runs for the Coastal
246 CHBr_3 tracer (Figure 5), with noticeably more CHBr_3 lofted into the TTL in the high
247 resolution run and with the peak value at a slightly higher altitude (Figure 5d). This feature,
248 with the peak at higher altitudes, is particularly prominent over the Maritime Continent. The
249 maximum in the CHBr_3 field on the 15.7 km surface does not coincide with the minimum in
250 potential temperature in either the coarse or high resolution runs, as might be expected
251 purely on the basis of the strength of convection. Rather the maximum tracer fields are
252 seen to the south and west of the main convection.

253 These features are similar when considered in potential temperature coordinates, as can
254 be seen from the potential temperature contours shown in Figures 4a and 5a. The highest
255 mixing ratios are found at levels between 365 and 370 K which are well above the main
256 level of zero net radiative heating; air at this level will be transported to the stratosphere
257 either vertically or along isentropic surfaces into the extratropical lower stratosphere.

258 When the emission sources are heterogeneous as is the case with the Coastal tracer
259 emissions, the magnitude of the vertical transport for a short-lived gas will depend on the
260 coincidence between source region and convective activity. In Section 4.1 we showed that
261 the HR run captures the strong convection over the Maritime Continent better than at the
262 coarser resolution (Figure 2). This strong convection combined with the large coastal
263 source around the Maritime Continent leads to the higher peak mixing ratios in bromoform,
264 at higher altitudes (Figure 5d), with enhanced transport likely into the stratosphere.

265 The representativeness of February 2005 can be examined by comparing the 5 years of
266 the high resolution run (1996, 1998, 2000, 2002, 2005) with (a) the same 5 years (Figure
267 6) and (b) all 10 years from the 10 year CR run (not shown). The same result was found
268 for each comparison. Figure 6 shows the vertical profiles (mean and variability) of the
269 CHBr_3 tracer for 4 cases: the Uniform tracer averaged over (a) the whole Tropics and (b)
270 the Maritime Continent; and (c) and (d) the Coastal tracer for the same two cases. With the
271 Uniform tracer emissions, the tropical average profiles and associated variability for CR
272 and HR shown in Fig 6(a) are similar. The variability is greater over the Maritime Continent
273 (Fig 6b), as might be expected for a region whose convection is strongly influenced by
274 ENSO events, though the difference between CR and HR versions is still small. The
275 variability found with the Coastal tracer is enhanced compared with the Uniform tracer in
276 both HR and CR runs. This enhancement is most noticeable over the Maritime Continent
277 where there is additionally a 10-15% increase in the peak CHBr_3 tracer amount in the high
278 resolution run, indicating that the effect in February 2005 is typical, though larger than in
279 other years.

280 It is also worth noting the small maximum in CHBr_3 at 5 km over the Maritime Continent.
281 This feature is present in the 5-year average as well as in 2005. We ascribe this to low
282 level convection around the coast-lines in the Maritime Continent as previously reported
283 for Borneo (Robinson et al., 2012). It is not apparent in the latitude band average.

284 **5. Discussion and Conclusions**

285 We use high and coarse resolution versions of the UKCA model to investigate the impact
286 that model resolution and the geographical distribution of emissions have on CHBr_3 mixing

287 ratios in the TTL. The study focuses on February 2005, with its representativeness
288 checked through comparisons with Februaries from other years. Comparing the OLR and
289 the precipitation from the two model runs with observations shows that the HR model
290 captures the convection more realistically than the CR run in terms of both strength and
291 location. We ascribe this difference mainly to the HR model's better description of the low
292 level circulation and sea breezes associated with the larger islands of the Maritime
293 Continent.

294 The HR model produces significant differences between how coastal and uniform
295 emissions affect the CHBr_3 mixing ratios in the TTL (Figure 3). The effect varies regionally
296 with, for example, over twice as much CHBr_3 over the Maritime Continent for the coastal
297 emission case. When averaged over the global TTL there is 15-20% more CHBr_3 in the
298 TTL with the Coastal emissions. Several of the CHBr_3 emission estimates currently used in
299 global models are based, at least in part, on aircraft measurements made in the free
300 troposphere and TTL (e.g., Warwick et al., 2006; Liang et al., 2010; Ordonez et al., 2012).
301 The inhomogeneity in Figure 3(b) shows that estimates based on aircraft measurements
302 are sensitive to (a) the location of the measurements; (b) the description of convection in
303 the model used; and (c) the assumed ratio of coastal and open ocean emissions. Many of
304 the aircraft measurements used to derive global emissions are located in or around the
305 Pacific Ocean (e.g., Liang et al., 2010; Navarro et al., 2015), where our calculations
306 indicate higher than average TTL mixing ratios for CHBr_3 . It seems likely that global
307 emission estimates based on these aircraft measurements could be biased high, which
308 could offer an explanation for some of the current discrepancies between the various
309 estimates (Carpenter, Reimann et al, 2014). The larger the relative contribution of coastal
310 emissions, the more important this factor will be. Conversely, aircraft measurements can
311 likely be used to improve regional (and hence global) emission estimates. Such a region-
312 by-region approach would probably require more measurements than our currently
313 available.

314 The transport of short-lived species into the TTL and on to the stratosphere depends on
315 the location of the emissions and on the location of the major vertical ascent occurring in
316 convection. A maximum flux into the TTL would occur when the region of emission and
317 convection exactly coincide. On the other hand, if emission is far from convection then it is
318 likely that substantial chemical loss could occur before any rapid vertical transport; the
319 overall flux into the TTL would then be low. Convection is modelled better at higher
320 resolution so the difference between TTL CHBr_3 calculated for the Uniform or Coastal
321 tracers can be large in some regions, as discussed above. For our CR integrations, the
322 difference in the global TTL mixing ratios of CHBr_3 due to emission distribution is smaller
323 (compare figure 4c, d with Figure 5c, d). Other models run at coarse resolution might also
324 be expected to underestimate the amount of short-lived tracer lifted to TTL regionally, with
325 the underestimation differing from year-to-year (Figure 6).

326 The differences are largest for short-lived species, and so the major effect on the
327 stratospheric Br budget will be felt through CHBr_3 with its tropical lifetime of ~15 days and
328 a potentially large proportion of emissions in coastal regions. Similarly, if iodine-containing
329 species play a role in upper tropospheric and stratospheric chemistry (Saiz-Lopez et al.,
330 2015), understanding their precise emission locations will be important and high resolution
331 modelling will be required to capture their local impact. On the other hand, the calculated
332 TTL mixing ratio of the other major short-lived contributor to stratospheric bromine,

333 dibromomethane (CH₂Br₂), will be relatively unaffected by the model resolution as it has a
334 ~3 month tropical lifetime (Carpenter et al., 2014) and dominant open ocean sources
335 (Ziska et al., 2013).

336 We have not examined differences in the impact on the stratosphere for the different
337 model resolutions and emission distributions. This would require a complete model
338 calculation, where the degradation products of bromoform are modelled in a fully
339 interactive chemistry scheme. We note that the multi-year averages of the CHBr₃ mixing
340 ratios in the global TTL are similar for the CR and HR models implying that the large-scale
341 performance of the two models is reasonably similar when the total emissions are the
342 same. This suggests that the resolution of the models currently used in multi-annual
343 integrations to study the transport of bromine into the global stratosphere is acceptable
344 (although unacceptable if the aim is to compare model results with observed chemical
345 distributions in the TTL or to infer emissions). However because the details of the
346 convection do change with resolution, any changes in the preferred transport pathways
347 with climate change may not be accurately modelled.

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513 Figure 1. Tracer emission fields of CHBr_3 used in the model runs described here: (a)
514 Uniform tracer and (b) Coastal tracer in in high resolution run; (c) Uniform tracer and (d)
515 Coastal tracer in coarse resolution run.

516 Figure 2. Observed and modelled fields for February 2005: (a) Outgoing Longwave
517 Radiation (OLR) from AIRS in W m^{-2} ; (b) precipitation from TRMM in mm day^{-1} ; (c) OLR
518 and (d) precipitation for the high resolution run; and (e) OLF and (f) precipitation for the
519 coarse resolution run.

520 Figure 3. The monthly average mixing ratios at 15.7 km in February 2005 are shown for (a)
521 the Uniform_50 tracer; and (b) the Coastal tracer. Average vertical profiles of the mixing
522 ratios for the two tracers are shown for (c) the Tropics (20°S - 20°N) and (d) the Maritime
523 Continent (20°S - 10°N ; 90°E - 160°E , as shown in (a) and (b)).

524 Figure 4. The monthly average mixing ratios for the Uniform tracer at 15.7 km in February
525 2005 are shown for (a) the high resolution run; and (b) the coarse resolution run. Contours
526 of monthly average potential temperature are shown in (a) and (b). Average vertical
527 profiles of the mixing ratios for the two runs are shown for (c) the Tropics (20°S - 20°N) and
528 (d) the Maritime Continent (20°S - 10°N ; 90°E - 160°E). High resolution run is shown in blue;
529 the coarse resolution run is shown in pink.

530 Figure 5. As for Figure 4, except that these plots are for the Coastal Tracer

531 Figure 6. Average vertical profiles of the mixing ratios for the Uniform tracer in five
532 representative Februaries (1996, 1998, 2000, 2002, 2005) are shown for (a) the Tropics
533 (20°S - 20°N) and (b) the Maritime Continent (20°S - 10°N ; 90°E - 160°E). Equivalent plots for
534 the Coastal tracer are shown in panels (c) and (d). High resolution run is shown in blue;
535 the coarse resolution run is shown in pink. Dashed lines indicate 2 standard deviations
536 from the mean (solid).

537