

1 **Three years of measurements of light-absorbing aerosols over coastal**

2 **Namibia: seasonality, origin, and transport**

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20

## 21 **Abstract**

22 Continuous measurements between July 2012 and December 2015 at the Henties Bay Aerosol  
23 Observatory (HBAO; 22°S, 14°05'E), Namibia, show that, during the austral wintertime,  
24 transport of light-absorbing black carbon aerosols occurs at low-level into the marine boundary  
25 layer. The average of daily concentrations of equivalent black carbon (eBC) over the whole  
26 sampling period is  $53 (\pm 55) \text{ ng m}^{-3}$ . Peak values above  $200 \text{ ng m}^{-3}$  and up to  $800 \text{ ng m}^{-3}$  occur  
27 seasonally from May to August, ahead of the dry season peak of biomass burning in southern  
28 Africa (August to October). Analysis of three-day air mass back-trajectories show that air  
29 masses from the south Atlantic ocean south of Henties Bay are generally cleaner than air having  
30 originated over the ocean north of Henties Bay, influenced by the outflow of the major biomass  
31 burning plume, and from the continent, where the wildfires occur. Additional episodic peak  
32 concentrations, even for oceanic transport, indicate that pollution from distant sources in South  
33 Africa and maritime traffic along the Atlantic ship tracks could be important. While we expect  
34 the direct radiative effect to be negligible, the indirect effect on the microphysical properties of  
35 the stratocumulus clouds and the deposition to the ocean could be significant and deserve  
36 further investigation, specifically ahead of the dry season.

## 37 **1. Introduction**

38 Aerosol particles of natural and anthropogenic origin affect the Earth's climate and modulate  
39 the greenhouse effect of long-lived gases (Boucher et al., 2013). The extent of this modulation  
40 depends on their nature, in particular on their chemical composition and size distribution  
41 determining their interactions with radiation and clouds. Current understanding suggests that  
42 atmospheric aerosols increase the global outgoing shortwave radiation, enhancing the  
43 atmospheric albedo, thereby counteracting the warming effect of greenhouse gases (Boucher et  
44 al., 2013). However, light-absorbing aerosols, such as black carbon (BC) from fossil fuel  
45 combustion and biomass burning, can reduce the amount of outgoing radiation at the top of

46 atmosphere (TOA), finally adding to the greenhouse effect (Haywood and Shine, 1995;  
47 Jacobson, 2001; Chung and Seinfeld, 2002; Bond and Bergstrom, 2006; Koch and Del Genio,  
48 2010; Bond et al., 2013). The heating radiative effect of black carbon aerosols is either enhanced  
49 or suppressed if they are above or below clouds, respectively (Keil and Haywood, 2003; Koch  
50 and Del Genio, 2010). The local heating induced by light-absorption below clouds could modify  
51 the cloud properties by enhancing the vertical motion and increasing the cloud cover and liquid  
52 water content (Koch and Del Genio, 2010). Finally, by entrainment into clouds, BC-containing  
53 aerosols could cause the cloud to evaporate and rise (Hansen et al., 1997) and reduce the cloud  
54 mean drop size diameters, increase droplet concentrations and henceforth reflectivity (Seinfeld  
55 and Pandis, 1997).

56 These processes are relevant to the western coast of southern Africa, pointed out by the latest  
57 Intergovernmental Panel for Climate Change (IPCC) report as a region where future warming  
58 and reductions in precipitation should be severe (Maure et al., 2018).

59 The west coast of southern Africa is characterised by a persistent and extended stratocumulus  
60 cloud deck topping a shallow, stable marine boundary layer maintained by the cold sea-surface  
61 temperatures of the Benguela Current (Cook et al., 2004; Tyson and Preston-Whyte, 2002), and  
62 by high loading of light-absorbing aerosols, mostly from seasonal biomass burning in the austral  
63 dry season (Swap et al., 2002), but possibly from various local and distant anthropogenic  
64 activities including ship traffic and energy production (Piketh et al., 1999; Formenti et al., 1999;  
65 Tournadre, 2004). Stratocumulus clouds are highly reflective and efficient in modifying the net  
66 radiative balance at TOA (Boucher et al., 2013). However, the mechanisms by which they could  
67 interact with light-absorbing aerosols, and the direct and indirect effects of those interactions  
68 on the regional radiative budget are largely unknown (Keil and Haywood, 2003; Flato et al.,  
69 2013; Myhre et al., 2013; Xu et al., 2014).

70 To address these questions, a large observational effort was initiated in the last few years by a  
71 number of coordinated intensive airborne and ground-based field campaigns, analysis of  
72 spaceborne observations, and climate modelling (Zuidema et al., 2016). These experiments  
73 focused on the dry season period between July and October, when biomass burning aerosols  
74 contribute by optically-dense plumes with instantaneous aerosol optical depth (AOD)  
75 systematically larger than 0.5 at mid-visible wavelengths (Swap et al., 2006). The emission,  
76 transport and direct radiative effect of light-absorbing carbonaceous aerosols by biomass  
77 burning aerosols also motivated previous experiments, such as the Southern African Regional  
78 Science Initiative (SAFARI 2000, Swap et al., 2006) and the Southern African Fire-Atmosphere  
79 Research Initiative (SAFARI, Andreae et al., 1996).

80 However, little is known about the aerosol concentrations and properties outside this season.

81 To fill this gap, this paper presents the first results of the mass concentrations of light-absorbing  
82 carbonaceous aerosols on the Atlantic coast of Namibia from three years of observations at the  
83 Henties Bay Aerosol Observatory (HBAO, 22°S, 14°05'E) long-term ground-based surface  
84 station.

85 Measurements of the mass concentrations of equivalent black carbon (eBC) recorded between  
86 July 2012 and December 2015 in the marine boundary layer below the stratocumulus deck are  
87 analysed to gather new knowledge on importance and seasonality. Observations are coupled  
88 with calculations of air mass back-trajectories to identify the dominant transport patterns and  
89 quantify their contributions. A comparison to the MERRA-2 model reanalysis is performed.

## 90 **2. Methods**

91 Surface observations of aerosol particles are conducted at the Henties Bay Aerosol Observatory  
92 (HBAO, [www.hbao.cnrs.fr](http://www.hbao.cnrs.fr)), a recent regional station in the Global Atmosphere Watch (GAW)  
93 Programme of the World Meteorological Organization (WMO). The research centre is located

94 on the Sam Nujoma Marine and Coastal Resources Research Centre (SANUMARC) of the  
95 University of Namibia in Henties Bay (22°S, 14°05'E), Namibia (Figure 1). Henties Bay is a  
96 small town in an arid environment with no vegetation, no industrial activity and very little  
97 traffic. Energy usage is predominantly a mix of electricity and gas, with some solid fuel  
98 combustion (wood) due to low availability (A. Namwoonde, 2017, *pers. comm.*). The  
99 monitoring site, situated on the University campus, is located on the coast approximately 100 m  
100 from the shore line. To the east are the Namibian Gravel Plains, at 3 km to the south of the  
101 campus is the town of Henties Bay, and to the north is the Omaruru Riverbed (river mouth  
102 approximately 100 m from SANUMARC). The population of Henties Bay ranges between 4  
103 600 and 6 000 inhabitants, according to the Namibia 2011 population and housing census (main  
104 report available at <http://cms.my.na/assets/documents/p19dmn58guram30ttun89rdrp1.pdf>).

## 105 **2.1. Measurements of light-optical attenuation**

106 Instruments at HBAO operate from a roof terrace at approximately 30 m above the ground. The  
107 terrace hosts the sampling inlets, from which air is drawn into a laboratory room located  
108 underneath by straight stainless-steel pipes to avoid particle losses. The optical attenuation of  
109 light (ATN) by aerosol particles smaller than 1  $\mu\text{m}$  in aerodynamic diameter was measured by  
110 a single-wavelength aethalometer (model AE-14U, Magee Sci., Berkeley, CA) operating at 880  
111 nm and sampling at 3.5 ( $\pm 0.1$ )  $\text{L min}^{-1}$  from a certified  $\text{PM}_{10}$  inlet (BGI Inc., Waltham, MA).  
112 The physical principle of operation of the aethalometer is detailed in Hansen et al. (1984).  
113 Measurements were performed at a 5-min time resolution and stored on a data logger (model  
114 CR-1000, Campbell Sci. Ltd.). The original data set was screened to eliminate spikes and peaks  
115 lasting less than two hours, generally associated with open fires for barbequing meat. The data  
116 record extended from July 2012 to December 2015, with an extended data gap between January  
117 and July 2014 due instrument maintenance.

118 The Lambert-Beer law relates the temporal variation of the measured light-attenuation (ATN)  
119 due to aerosol particles collected on a quartz fibre tape to the mass concentration of eBC (in  $\mu\text{g}$   
120  $\text{m}^{-3}$ ). This is based on the fact that black carbon is the strongest light-absorber in the near  
121 infrared (Kirchstetter et al., 2004; Caponi et al., 2017).

122 The operational equation linking eBC to the attenuation (ATN) measured by the aethalometer  
123 is

124

$$125 \quad \text{eBC} = \frac{1}{\text{MAC}_{\text{BC}}} \left( \frac{1}{C \cdot R(\text{ATN})} \right) \left( \frac{A \Delta \text{ATN}}{V \Delta t} \right) \quad (1)$$

126

127 where  $A$  represents the area of the aerosol deposit on the filter,  $V$  the volumetric flow rate, and  
128  $\Delta \text{ATN}/\Delta t$  is the variation rate of attenuation with time. The terms  $C$  and  $R(\text{ATN})$  account for  
129 measurement artefacts that artificially increase absorption estimated from attenuation  
130 measurements. The term  $C$  takes into account the multiple scattering effects on the filter due to  
131 both the filter fibers and the aerosol particles embedded in it. The factor  $R(\text{ATN})$  accounts for  
132 the shadowing effect occurring with time as high concentrations of absorbing particles are  
133 collected on the filter. Published values of the  $C$  parameter at 660 and 880 nm range between  
134 1.75 and 6.3 depending on the nature of the light-absorbing aerosols, the measurement  
135 environment, and finally on the parametrisation of the corrections (Weingartner et al., 2003;  
136 Arnott et al., 2005; Schmid et al., 2006; Collaud-Coen et al., 2010; Segura et al., 2014; Saturno  
137 et al., 2017; Di Biagio et al., 2018). These authors show that, regardless of location, values  
138 below 3.5 are appropriate for moderately absorbing aerosols whose single scattering albedo  
139 ( $\omega_0$ ) is above 0.8 at 660 nm. For Mace Head, a coastal site with prevailing marine North Atlantic  
140 air masses, Collaud-Coen et al. (2010) reported a mean  $C$  value of 3.44 ( $\pm 0.21$ ), which we used  
141 for HBAO, neglecting any possible wavelength dependence. The parametrisation of the

142 shadowing effect  $R(ATN)$  depends on  $\omega_0$ , henceforth, on the availability of concurrent  
143 measurements of the scattering coefficient. This is the case at HBAO, where, however,  
144 scattering is measured on the  $PM_{10}$  and not on the  $PM_1$  fraction as attenuation is, preventing a  
145 meaningful estimate of  $\omega_0$ . In this case, Collaud-Coen et al. (2010) recommended the  
146 Weingartner et al. (2003) correction, leading to a mean value of the R parameter of 0.93, which  
147 we assumed for the further analysis.

148 The other crucial parameter in Equation (1) is the mass absorption efficiency of eBC ( $MAC_{BC}$ ,  
149 units of  $m^2 g^{-1}$ ). Many authors have reported values in the range 5-20  $m^2 g^{-1}$  at wavelengths  
150 between 550 and 870 nm, and related this variability to the chemical state and age of black  
151 carbon aerosols (Liousse et al., 1993; Petzold et al., 1997; Martins et al., 1998; Kirchstetter et  
152 al., 2003; Hansen, 2005; Bond and Bergstrom, 2006; Knox et al., 2009; Subramanian et al.,  
153 2010; Bond et al., 2013; Zanatta et al., 2016). More recently, Zuidema et al. (2018) reported  
154 that the  $MAC_{BC}$  at 648 nm at Ascension Island, farther west than HBAO, and at times in its  
155 outflow, varied between 14.1  $m^2 g^{-1}$  in June to 10.7  $m^2 g^{-1}$  in July to October. When  
156 extrapolated, these values result in a  $MAC_{BC}$  at 870 nm between 7.9 and 10  $m^2 g^{-1}$ . Their  
157 average and standard deviation ( $9.0 \pm 1.5 m^2 g^{-1}$ ) was retained in our analysis.

## 158 **2.2. Supporting data**

159 In 2013, the mass concentration of particles of diameter smaller than 2.5  $\mu m$  in equivalent  
160 aerodynamic diameter ( $PM_{2.5}$ ) was sampled by a Tapering Element Oscillating Microbalance  
161 (TEOM, model 1400a, Rupprecht and Patashnick, Albany, New York, USA) operating from a  
162 certified  $PM_{2.5}$  inlet (also from Rupprecht and Patashnick). The total flow rate at the inlet was  
163 16.7  $L min^{-1}$  to ensure the correct functioning of the inlet, and the sampling flow rate driving  
164 the aerosol-laden air to the microbalance was 3  $L min^{-1}$ . The temperature of the sample stream  
165 was kept constant at 50°C.

166 Three-dimensional air mass back-trajectories are calculated using the NOAA HYbrid Single-  
167 Particle Lagrangian Integrated Trajectory Model (HYSPLIT; Draxler and Rolph, 2015). The  
168 model uses the  $1^{\circ} \times 1^{\circ}$  latitude-longitude grid, reanalysis meteorological database. The 6-hourly  
169 reanalysis archive data are generated by the NCEP's GDAS (NCEP: National Centers for  
170 Environmental Prediction; GDAS: Global Data Assimilation System) wind field reanalysis.  
171 Further information can be found at <https://rda.ucar.edu/datasets/ds083.2/>.

172 ERA-Interim reanalysis data (Dee, et al. 2011) from the European Center for Medium Range  
173 Weather Forecast (ECMWF) are used in the analysis of synoptic scale circulation patterns  
174 associated with the identified dominant air mass transport to Henties Bay. The 6 hourly (0, 6,  
175 12, 18 UTC) analysis data ( $0.75 \times 0.75$  degrees) at mean sea level pressure (MSLP - variable  
176 151.128) and the 500 hPa geopotential height (ZG500 -- variable 129.128) are used for this  
177 study. Data sets were normalised (MSLP / 100 and ZG500 /100) using Climate Data Operators  
178 (CDO) (Schulzweida et al., 2006) and plotted with 2 hPa and 10 hPa isobaric intervals for  
179 MSLP and 500 hPa level, respectively.

180 Surface black carbon concentrations predicted by the Modern-Era Retrospective analysis for  
181 Research and Applications, Version 2 (MERRA-2, Gelaro et al., 2017), sampled at HBAO and  
182 at a number of other sites (Zuidema et al., 2016) are used for comparison.

183 The HTAP\_V2 dataset is used for gridded emission of anthropogenic black carbon for the year  
184 2010 (Janssens-Maenhout et al., 2015). It consists of  $0.1^{\circ} \times 0.1^{\circ}$  grid-maps. HTAP\_V2 uses  
185 nationally reported emissions combined with regional scientific inventories in the format of  
186 sector-specific grid maps. The grid maps are complemented with EDGARv4.3 data for those  
187 regions where data are absent. Anthropogenic activities producing black carbon aerosols  
188 comprise aviation, transportation, energy production, industries, ship traffic, residential and  
189 agricultural burning.



## 190 **3. Results**

### 191 **3.1. Temporal variability of eBC concentrations**

192 Figure 2 shows daily and monthly averages of the eBC concentrations measured at HBAO  
193 between July 2012 and December 2015. Daily averages excluded spikes and peak values  
194 occurring on short time scales, less than 1-2 hours, resulting from contamination of local  
195 activities (episodic traffic and occasional open fires for barbequing meat).

196 The daily mean average of  $53 (\pm 55) \text{ ng m}^{-3}$  is in accordance with previous observations in  
197 remote locations of the world shown in Table 1 (Bodhaine, 1995; Andreae et al., 1995; Derwent  
198 et al., 2001; von Schneidemesser et al., 2009; Marinoni et al., 2010; Sheridan et al., 2016).  
199 Andreae et al. (1995) found eBC mass concentrations lower than  $50 \text{ ng m}^{-3}$  along a cruise  
200 transect at  $19^\circ\text{S}$  over the south east Atlantic between Brazil and Angola, except when  
201 approaching the African continent, when concentrations increased in the range  $50\text{-}150 \text{ ng m}^{-3}$ ,  
202 indicating a strong continental influence in this otherwise pristine environment. Additional  
203 published research, also in Table 1, reports absorption coefficients that would lead to  
204 comparable eBC concentrations (Bodhaine, 1995; Clarke, 1989; Quinn et al., 1998). For  
205 contrast, eBC mass concentrations in lofted layers above the marine boundary layer in the range  
206  $0.1\text{-}6 \mu\text{g m}^{-3}$  were reported for aged biomass burning haze (Kirchstetter et al., 2003; Formenti  
207 et al., 2003; Eatough et al; 2003), and up to  $5\text{-}40 \mu\text{g m}^{-3}$  for fresh biomass smoke plumes  
208 (Kirchstetter et al., 2003).

209 Figure 2 also shows an apparent seasonal variability in eBC, further highlighted by the monthly  
210 means and by the comparison with  $\text{PM}_{2.5}$  mass concentration measurements performed at the  
211 site during 2013, which conversely, did not display any particular seasonal cycle (Figure S1),  
212 likely because dominated by sea salt. Concentrations increase in the austral winter from May  
213 to July, and decrease from August to April. The increase from May to July is well captured by  
214 the MERRA-2 reanalysis (also shown in Figure 2), according to which, however,

215 concentrations only start decreasing after September. The observed seasonality is somewhat  
216 surprising in that it precedes the seasonal maximum of the biomass burning fire season in  
217 southern Africa, peaking in the austral dry season from August to October (Swap et al., 2002).  
218 As previously stated in this paragraph, data constituting the time series have been screened to  
219 exclude short-term variability (less than one-two hour time intervals) to exclude isolated and  
220 episodic sources. Peaks of eBC driving the seasonal increase in the May-to-August period are  
221 long-lasting, extending between 6 and 11 hours, and occurring during both day- and night-time.  
222 This suggests that transport is the cause of the seasonal peaks. The following section explores  
223 this hypothesis and attempts the quantification of attribution of eBC peaks to specific transport  
224 patterns.

### 225 **3.2. General atmospheric circulation driving air mass transport**

226 Transport to the west coast of Namibia is influenced by four general circulation patterns:  
227 baroclinic westerly waves, barotropic easterly waves, the semi-permanent south Atlantic high  
228 pressure and a continental high pressure circulation. The relative influence of each circulation  
229 pattern is highly seasonal and driven by the meridional migration towards the north in austral  
230 winter and to the south in summer. The origin of the air parcel over the southern ocean is linked  
231 to the passage of a westerly wave and front that propagates towards the subcontinent from the  
232 south west. These are Rossby waves that form as a result of the extratropical temperature  
233 gradient with a maximum impact on the weather over southern Africa in winter. The easterly  
234 waves are trade winds that are associated with the position of the Inter Tropical Convergence  
235 Zone (ITCZ) that reaches a maximum over the subcontinent during summer (Tyson and  
236 Preston-Whyte, 2014; Taljaard, 1994). The semi-permanent high pressure system (anticyclone)  
237 results from the descending limb of the Hadley circulation that interacts with the  
238 aforementioned waves. The south Atlantic high pressure system will ridges behind a passing  
239 westerly wave in the direction of maximum cold air advection. Conversely, a strengthening

240 anticyclone will block propagations of these waves and induce a strong persistent continental  
241 high pressure. Air masses reaching the sampling station have been found to originate from the  
242 adjacent Atlantic Ocean and various locations over the continental subcontinent. Coastal lows  
243 are induced along the west coast of Namibia and result in offshore flow ahead of westerly  
244 waves. This low pressure system forms localised cyclonic circulation that includes onshore flow  
245 in the north and offshore flow in the south of the low pressure cell (Tlhalerwa et al., 2005;  
246 Tyson and Preston-Whyte, 2014).

### 247 **3.3. Identification of air mass transport pathways impacting HBAO**

248 Three-day back-trajectories calculated daily between July 2012 and December 2015 are  
249 grouped according to the progression of the general synoptic circulation patterns and assigned  
250 to 8 geographical sectors according to the position of their end point, shown in Figure 3. Four  
251 sectors (G1 to G4) correspond to oceanic air masses and sectors (G5 to G7) to transport from  
252 the continent. The last sector (G8) describes air masses recirculating around the sampling site  
253 for most of the three day period. Figure 3 also shows the 2010 black carbon aerosol regional  
254 emission HTAP\_V2 inventory grid map from anthropogenic activities. Emissions are low in  
255 Namibia and neighbouring countries as Botswana and the west-central South Africa. Areas of  
256 higher emissions are Angola (with an hotspot in correspondence with the capital city Luanda),  
257 costal South Africa, particularly to the east, but also to the south in the Cape Town greater area,  
258 but mostly in the South African Highveld (27°S, 28°W) where the energy production is  
259 concentrated. The open-ocean and coastal ship tracks are also evident.

260 The monthly distribution of fire counts from 2012 to 2015 provided by MODIS/Aqua is shown  
261 in Figure 4. Although some interannual variability exists, the image record is consistent in  
262 showing that the fire season in Southern African starts towards April and extends until October.  
263 The major source areas are north of Namibia (Angola, Zambia), in South Africa (to the east and

264 along the south coast) and in Mozambique. In Namibia, fire counts are seen towards the north,  
265 around the Etosha Pan desert.

266 The seasonal contribution of these air masses transport pathways is shown in Figure 5. Sectors  
267 G1 to G4 represent the most common air flow pathway (73% out of 1279 calculated back-  
268 trajectories). The Southern Atlantic Ocean transport (G1 and G2) is the dominant surface  
269 circulation along the west coast, resulting from the northward moving limb of the surface south  
270 Atlantic high pressure. Air masses originate over the southern Atlantic Ocean, as far south as  
271 55°S (sector G2, representing approximately 66% of the air mass occurrences). During summer  
272 this is predominantly a function of the most southerly location of the centre of the south Atlantic  
273 high (Figure 6). In winter transport results from a complex interaction between the westerly  
274 waves propagating from the south west over the subcontinent and the reestablishment of the  
275 south Atlantic High in the westerly waves wake. Initially, air transport is towards the east and  
276 is then directed northwards along the west coast to Namibia (Figure 7). The distance covered  
277 by these air masses is several thousand kilometres due to the high wind speeds associated with  
278 the initial transport in the cyclonic circulation.

279 Sectors G3 and G4 describe transport from the tropical regions of the Atlantic Ocean. The  
280 onshore flow towards the sampling site forms as a westerly wave advances. A shallow, localised  
281 cell of low pressure (cyclonic circulation) is induced along the west coast with a diameter of  
282 approximately 200 km. Towards the north of the cell onshore flow occurs, while in the southern  
283 portion of the cell the flow is offshore. This circulation has also been shown to induce the dust  
284 plumes that blow off the Namibian coast over the Atlantic Ocean from ephemeral river beds  
285 along the west coast of Namibia (Tlhalerwa et al., 2005). The low, referred to as a coastal low,  
286 then propagates southwards behind the surface front. It is possible for air that is moved offshore  
287 in the easterly wave over Angola to be caught up in this more near shore circulation.

288 Transport to HBAO from the continent (sectors G5-G7) occurred on 19% of the total days  
289 (sectors G5, G6 and G7). These transport pathways are directly linked to the position of the  
290 easterly wave over the subcontinent as well as the position of the trough line associated with  
291 the wave. As pointed out earlier, the position of the easterly wave is highly seasonal. In general,  
292 air is transported across the subcontinent and exits to the Atlantic Ocean in the westerly  
293 transport. The low pressure trough moves across the subcontinent. The position of the trough  
294 also determines the exact pathway of transport as well as the sector in which air masses  
295 originate. If the trough is situated along the west coast it forms a west coast trough that  
296 facilitates flow along and close to the west coast. During summer the easterly wave reaches to  
297 the southern tip of southern Africa. This leads to transport of air from areas of South Africa,  
298 including the highly industrialised Highveld region (Figure 8a). In winter the easterly wave  
299 seldom reaches south of 25°S. Air masses during this season are more likely to originate over  
300 the central portion of southern Africa (Figure 8b).

301 Finally, sector G8 is associated with air masses originating within 100 km of HBAO (Figure  
302 9), either from land or from the ocean, and representing about 8% of the air mass occurrences  
303 (Figure 5). This circulation, only occurring in the second half of each year (Figure 5), is linked  
304 to the formation of a low pressure heat cell close to the west coast of Namibia centred at about  
305 the latitude of Henties Bay. Despite this being cyclonic flow the circulation is closed (Figure  
306 9) and therefore represents transport from close to the sampling site. The heat low is always  
307 embedded in an easterly wave or west coast trough. Centres of low pressure form along the  
308 west coast producing local and mesoscale circulation from the interior of Namibia to the coast.  
309 This flow pattern is distinguishable from a coastal low as it is centred on the subcontinent  
310 whereas the coastal low is always centred on the coast just offshore.

### 311 **3.4. Contribution of air transport patterns to the measured eBC**

312 Figure 10 illustrates the contribution of the air mass sectors G1-G8 to the eBC mass  
313 concentrations measured at HBAO and those estimated by the MERRA-2 reanalysis. This has  
314 been done by calculating the distribution of eBC values per group.

315 Although the absolute values differ by a factor of 2-3, measurements and reanalysis show some  
316 consistent temporal features. Episodic high values of eBC concentrations occur independently  
317 on the origin of the air mass. The southern Atlantic oceanic air masses (sectors G1, G2 and G3)  
318 and the continental G7 sector, corresponding to the low population density semi-arid region of  
319 the Karoo, in South Africa, display the lowest concentrations. In particular, the oceanic sectors  
320 are characterised by a south-to-north gradient, the highest mean concentrations being from  
321 sectors G4, offshore northern Namibia and Angola, comparable to those from the continental  
322 sectors G5 and G6, and G8, representing recirculating air masses. Measurements at HBAO  
323 indicate that the contributions of sectors G5 and G6 are equivalent, while sector G5 is the largest  
324 contributor according to the MERRA-2 reanalysis.

### 325 **4. Discussion and conclusions**

326 This papers present the first long-term time series of equivalent black carbon concentrations in  
327 the marine boundary layer on the south-east Atlantic coast offshore southern Africa.  
328 Observations were conducted at the Henties Bay Aerosol Observatory, in Namibia, between  
329 July 2012 and December 2015.

330 Higher concentrations of eBC on the western coast of southern Africa are observed from April  
331 to July within continental and marine air masses north of 30°S (sectors G4, G5 and G6). Daily  
332 eBC peak concentrations at HBAO do not exceed 800 ng m<sup>-3</sup>, and are seldom larger than 200  
333 ng m<sup>-3</sup>, lower than measured at Ascension Island, approximately 1500 kilometres downwind of  
334 coastal Namibia and located along the main outflow pathway from southern Africa to the

335 Atlantic Ocean (Swap et al., 1996; 2002; Adebisi and Zuidema, 2016; Zuidema et al., 2018).  
336 The seasonality of the eBC concentrations observed at HBAO corresponds to the seasonal shift  
337 from southern to northern circulation at the surface, and is in phase with the April onset of the  
338 fire season in southern Africa (Figure 5). The seasonal increase at HBAO is also well captured  
339 by the MERRA-2 reanalysis model, but it occurs earlier than reported by Zuidema et al. (2018)  
340 at Ascension Island (June to August). This seems to indicate that HBAO is on a minor branch  
341 of the transport pathway of the continental biomass burning smoke plume from continental  
342 southern Africa compared with the biomass burning plumes that reach Ascension Island. The  
343 MERRA-2 reanalysis shows higher concentrations than measured at HBAO and suggests that  
344 the period of high concentrations should persist until September rather than August as in the  
345 HBAO measurements. This points out to the inherent degree of uncertainty in our estimates.  
346 The correction factors (filter loading and multiple scattering corrections) needed to convert the  
347 measured attenuation into a value eBC concentration are assumed and not evaluated from  
348 concurrent measurements, and set to fixed values as the aerosol at HBAO would derive from a  
349 single source type. We do not deal either with potential changes of the aerosol properties due  
350 to ageing. Differences could also be due to the representation of the timing and extent of a  
351 southward shift of the easterly during summer. Although these issues cannot be resolved with  
352 the present dataset, they question the representation of the transport of smoke plumes at the  
353 subcontinental scale of southern Africa.

354 There is no doubt that the transport of wildfire smoke is the major regional source of the eBC  
355 aerosols for the western coast of Namibia. However, the presence of episodic outliers and the  
356 relatively elevated concentrations observed for oceanic air masses originating south of HBAO  
357 (sectors G1 to G3 in Figure 7) suggests that additional sources could contribute to the load of  
358 light-absorbing aerosols in the marine boundary layer. In particular, the contribution of the  
359 coastal and open ocean maritime shipping routes in the south Atlantic ocean (Tournadre, 2014;

360 Fraser et al., 2016; Johannson et al., 2017), and that of long-range continental anti-cyclonic  
361 transport from the industrial areas of the South African Highveld, showing up in Figure 3  
362 (Piketh et al., 2002), should be further explored.

363 By the very rough assumption of the mass fraction of black carbon to the total fine aerosol  
364 (10%, Bond et al., 2013), we estimate that the mean fine mass of aerosols containing eBC would  
365 be  $0.5 (\pm 0.5) \mu\text{g m}^{-3}$ . For comparison, the mean  $\text{PM}_{2.5}$  mass concentration at HBAO was  $14 (\pm$   
366  $11) \mu\text{g m}^{-3}$  in 2013 (Figure S1).

367 These aerosols below clouds would have a negligible direct radiative effect. There are almost  
368 no AERONET measurements of the aerosol optical depth (AOD) at HBAO concurrent to the  
369 eBC data series. However, Figure S2 shows the time series of the AERONET level 2.0 AOD  
370 of the fine and coarse mode aerosols ( $\text{AOD}_F$  and  $\text{AOD}_C$ ) evaluated by the O'Neill et al. (2003)  
371 algorithm between December 2011 and May 2012, and then from May to December 2015.  
372 Figure S2 shows that the  $\text{AOD}_F$  varies significantly from background values in the December  
373 2011-May 2012 period (average  $0.05 \pm 0.03$ ) to peak values of 0.4 and higher during August,  
374 September and October 2015, when the transport of biomass burning occurs in the free  
375 troposphere (Swap et al., 2003). The  $\text{AOD}_C$ , contributed essentially by sea salt, is relatively  
376 invariant with time. There is no process other than biomass burning that would inject aerosols  
377 above the marine boundary layer, henceforth we can consider the mean value for December  
378 2011-May 2012 as a reasonable evaluation of the optical depth of the fine mode of aerosols  
379 below clouds, including eBC.

380 The eBC aerosols might act on the microphysical properties of the local stratocumulus clouds.  
381 At Ascension Island, Zuidema et al. (2018) demonstrated the good correlation between the  
382 concentrations of refractory black carbon and cloud condensation nuclei (CCN) at  
383 supersaturations exceeding 0.2%. A similar effect should be expected at HBAO and could be  
384 important, outside but also during the biomass burning season as the entrainment of biomass



385 burning aerosols from the free troposphere maybe be inhibited by the thermal inversions clear  
386 air slots separating the elevated plumes and the marine boundary layer (Keil and Haywood,  
387 2003; Haywood et al., 2003; Hobbs et al., 2003). Finally, by deposition, these low-level aerosols  
388 could act on the biological activity for the oligotrophic south Atlantic gyre in summer,  
389 providing with soluble nutrient species, such as dissolved nitrogen, phosphorous, and iron  
390 (Guieu et al., 2005; Luo et al., 2008; Paris et al., 2010).

391 In conclusion, the chemical apportionment of the AOD<sub>F</sub> below cloud and the hygroscopic  
392 properties of the eBC aerosols at HBAO deserves exploration by future refined experiments.

### 393 **Data availability**

394 Original data for measured light-attenuation at HBAO are distributed by the French national  
395 AERIS data center (<https://www.aeris-data.fr/direct-access-icare-2/>). Treated data can be  
396 obtained by email request to the first author of this paper.

### 397 **Author contributions**

398 Paola Formenti, Stuart John Piketh, Andreas Namwoonde, Samuel Mafwila, Edosa Omoregie,  
399 Wolfgang Yunkermann and Willy Maenhaut designed the experiments and the sampling site.

400 Paola Formenti, Stuart John Piketh, Andreas Namwoonde, Mathieu Cazaunau, Anaïs Feron,  
401 Cécile Gaimoz, Stephen Broccardo, Nicola Walton, Karine Desboeufs, and Mattheus  
402 Hanghome performed the experiments.

403 Paola Formenti and Stuart John Piketh performed the full data analysis with contributions by  
404 Danitza Klopper, Guillaume Siour, and Roelof Burger.

405 Paola Formenti, Stuart Piketh, Danitza Klopper and Roelof Burger wrote the paper with  
406 comments from all co-authors.

### 407 **Competing interests**

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

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424 first place the scientific community for hemispheric transport of air pollution. The static version  
425 is available on this EDGAR website, but also the GEIA data portal and the ECCAD server.  
426 ECCAD is the GEIA Global Emission Initiative database ([www.geiacenter.org](http://www.geiacenter.org)) and is part of  
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670 **Table captions**

671 **Table 1.** Values of mass concentrations of equivalent black carbon (eBC) from measurements  
672 published in the literature for remote regions worldwide. When available, the specific  
673 attenuation used to convert the measured attenuation to eBC is also reported.

674 **Figure captions**

675 **Figure 1:** Geographical location of the Henties Bay Aerosol Observatory (HBAO).

676 **Figure 2.** Top panel (a): comparisons of the time series of daily eBC mass concentrations (ng  
677 m<sup>-3</sup>) measured at HBAO and predicted by the MERRA-2 reanalysis. The light grey boxes  
678 indicate periods of increasing concentrations. The light blue boxes indicate periods of  
679 decreasing concentrations. Bottom panel (b) box and whisker plot representation of the  
680 respective monthly variability.

681 **Figure 3.** Geographical boundaries of the sectors used to classify the air mass back trajectories  
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683 anthropogenic activities for the year 2010 provided by the HTAP\_V2 inventory. Emissions are  
684 expressed in Tons.

685 **Figure 4.** Fire counts per pixels from MODIS/Aqua provided by the NASA Fire Information  
686 for Resources Management System (FIRMS). Colours range from yellow (1 fire count per  
687 pixel) to red (+100 fire count per pixel). The underlying image is the corrected reflectance (true  
688 colour) measured by MODIS/Aqua on the first day of each months.

689 **Figure 5.** Seasonal variation in the transport pathways of air masses reaching HBAO between  
690 2012 and 2015.

691 **Figure 6.** Case study of mean sea level pressure over the sub-continent and adjacent South  
692 Atlantic Ocean for 16-19 November 2014 illustrating the synoptic circulation that results in the  
693 transport of air masses from sector G1.

694 **Figure 7.** Case study of mean sea level pressure over the sub-continent and adjacent South  
695 Atlantic Ocean for 1-4 June 3013 illustrating the synoptic circulation that results in the  
696 transport of air masses from sector G2.

697 **Figure 8.** Case study of mean sea level pressure over the sub-continent and adjacent south  
698 Atlantic ocean for A) summer (10-13 December 2013) and B) winter (6-10 July 2013)  
699 illustrating the synoptic circulation that results in the transport of air masses from sector G5-  
700 G7.

701 **Figure 9.** Case study of mean sea level pressure over the sub-continent and adjacent South  
702 Atlantic Ocean for 13-16 December 2012 illustrating the synoptic circulation that results in the  
703 transport of air masses from sector G8.

704 **Figure 10.** Contribution of air mass sectors to the eBC concentrations at HBAO from (a) our  
705 measurements and (b) MERRA-2 reanalysis.

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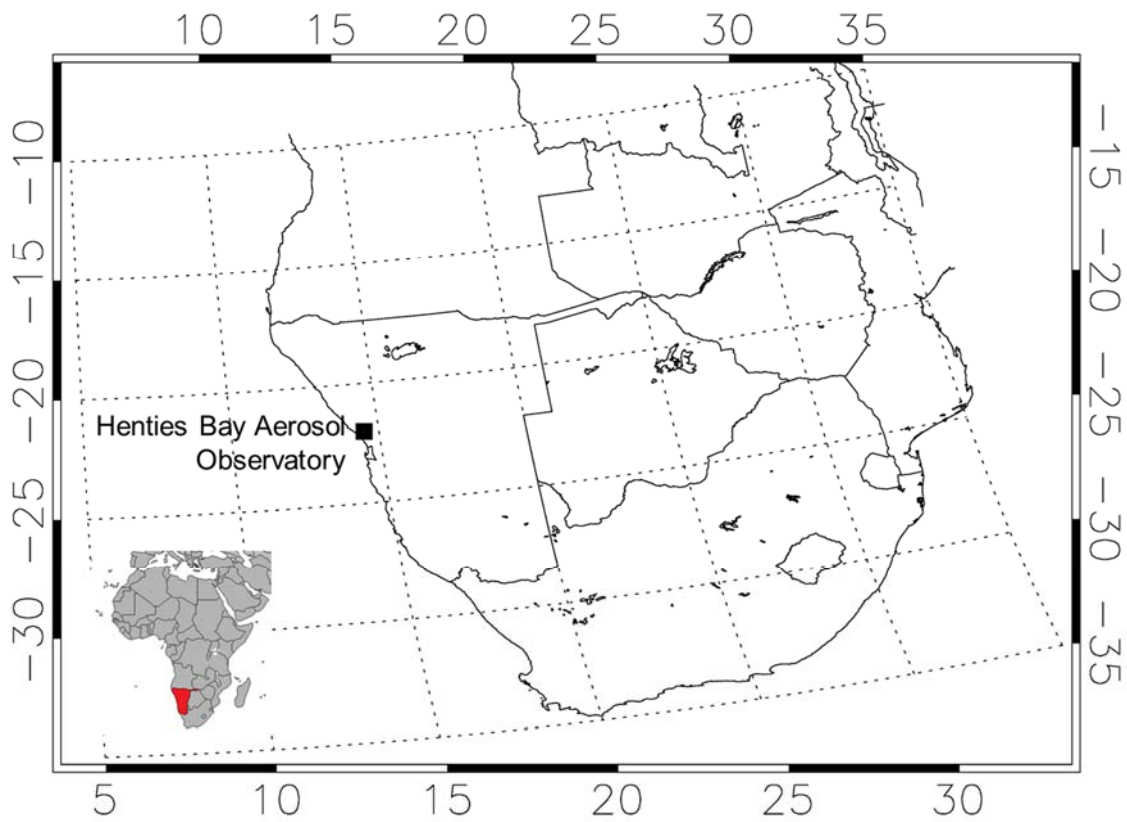
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 709 attenuation  $\sigma_{BC}$  used to convert the measured attenuation to eBC is also reported.

710

Location	eBC (ng m <sup>-3</sup> )	$\sigma_{BC}$ , 880 nm (m <sup>2</sup> g <sup>-1</sup> )	Reference
Tropical South Atlantic off southern Africa, 19°S	50-150	10	Andreae et al. (1995)
Nepal Climate Observatory Pyramid, Himalaya	160 ± 296	6.5 <sup>s</sup>	Marinoni et al. (2010)
Summit, Greenland	<340	-----	von Schneidmesser et al. (2009)
Mace-Head, Ireland	47-74	11 ± 3	Derwent et al. (2001)
South Pole, Antarctica	0.01-50	19	Bodhaine (1995)

711 <sup>s</sup> At 635 nm – measurements were conducted with a Multi-Angle Absorption Photometer  
 712 (MAAP 5012, Thermo Electron Corporation).  
 713

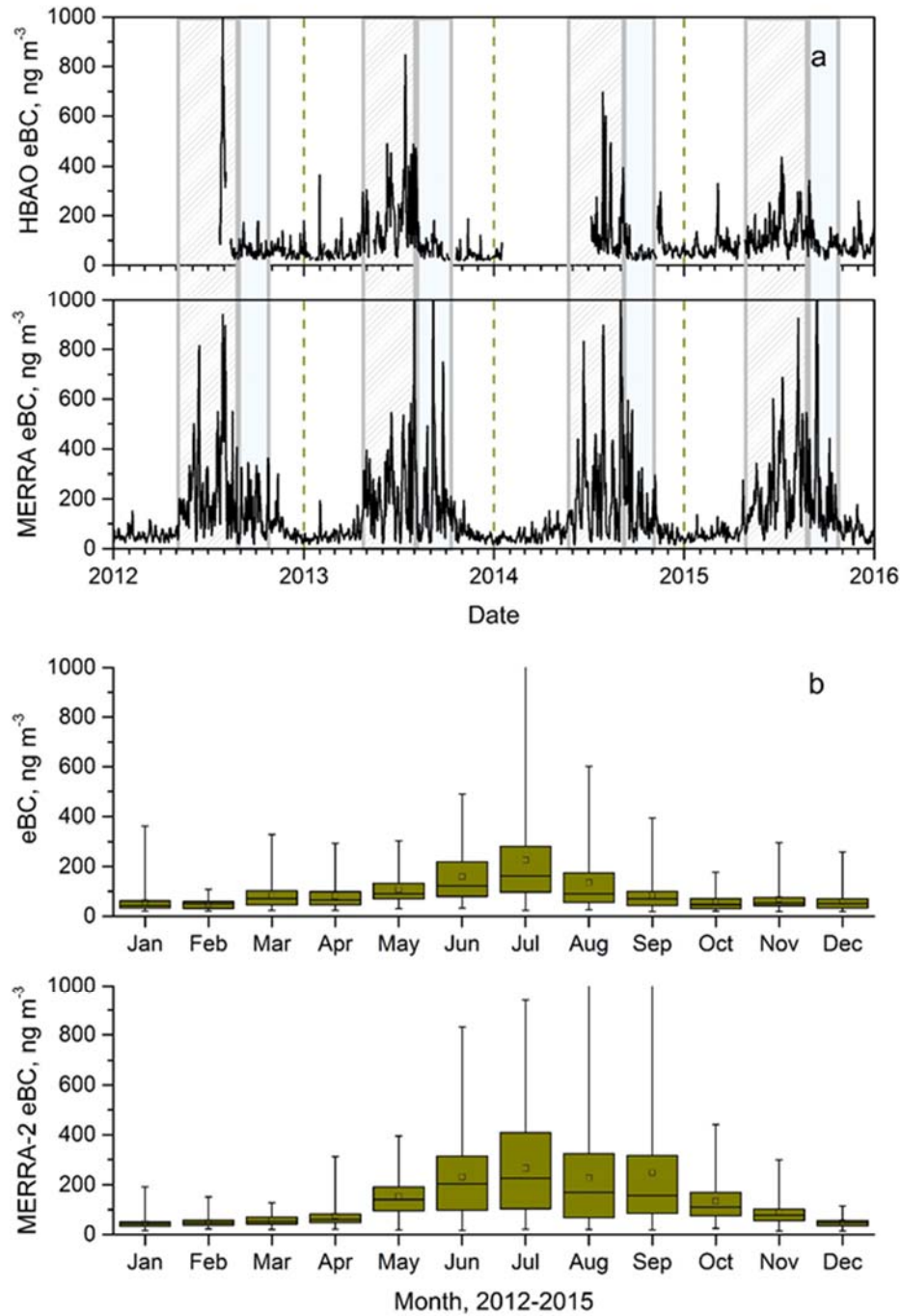
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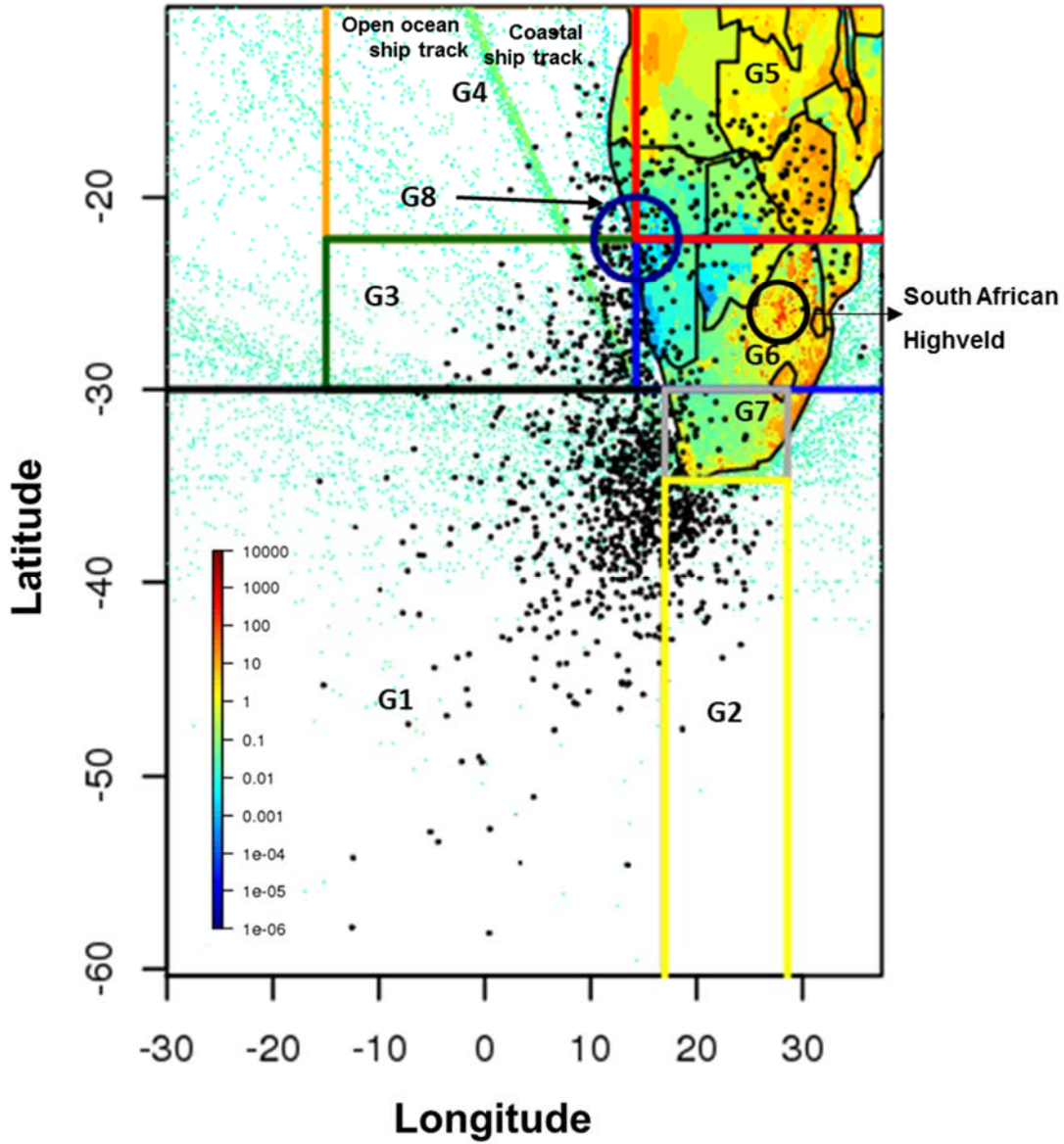
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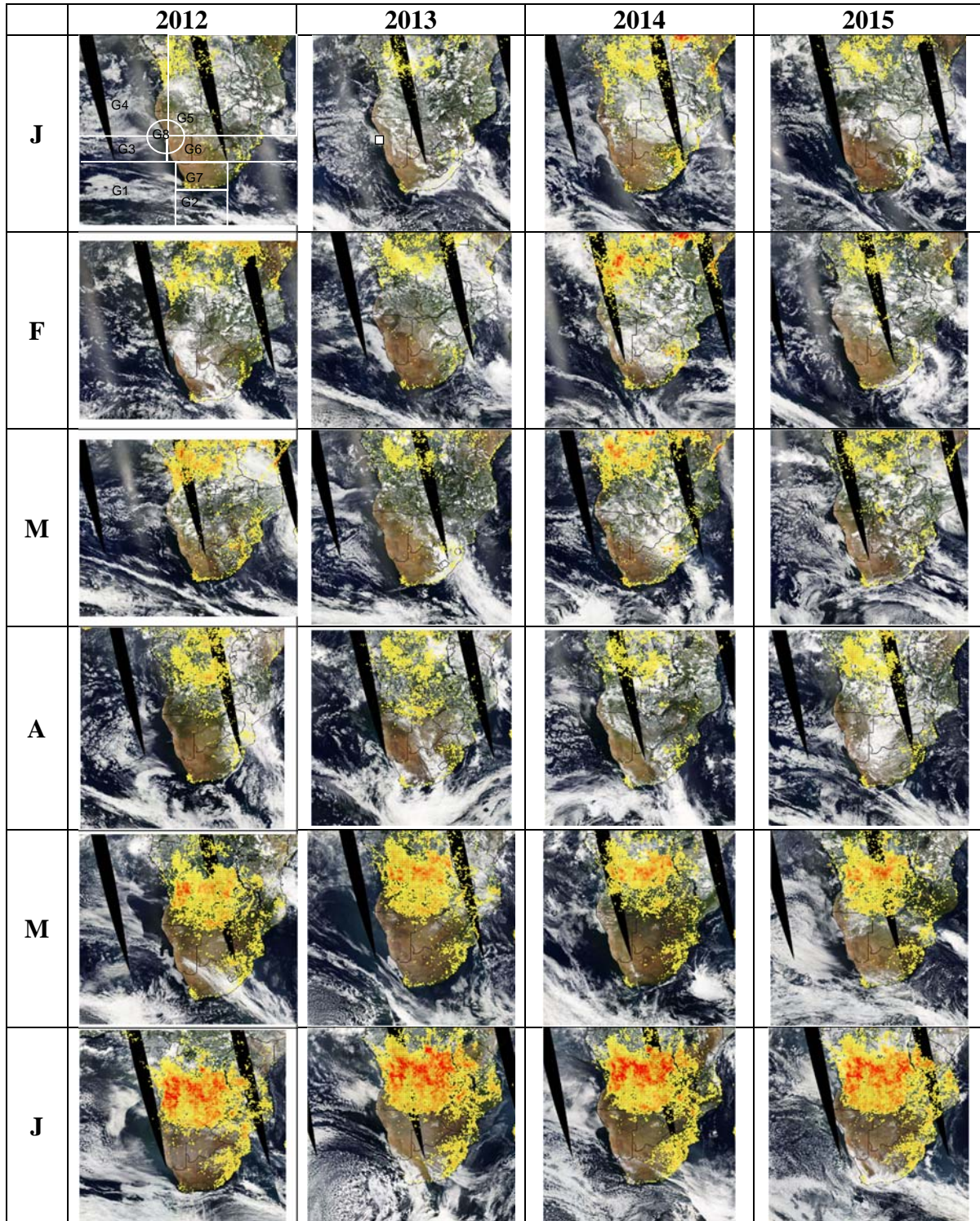
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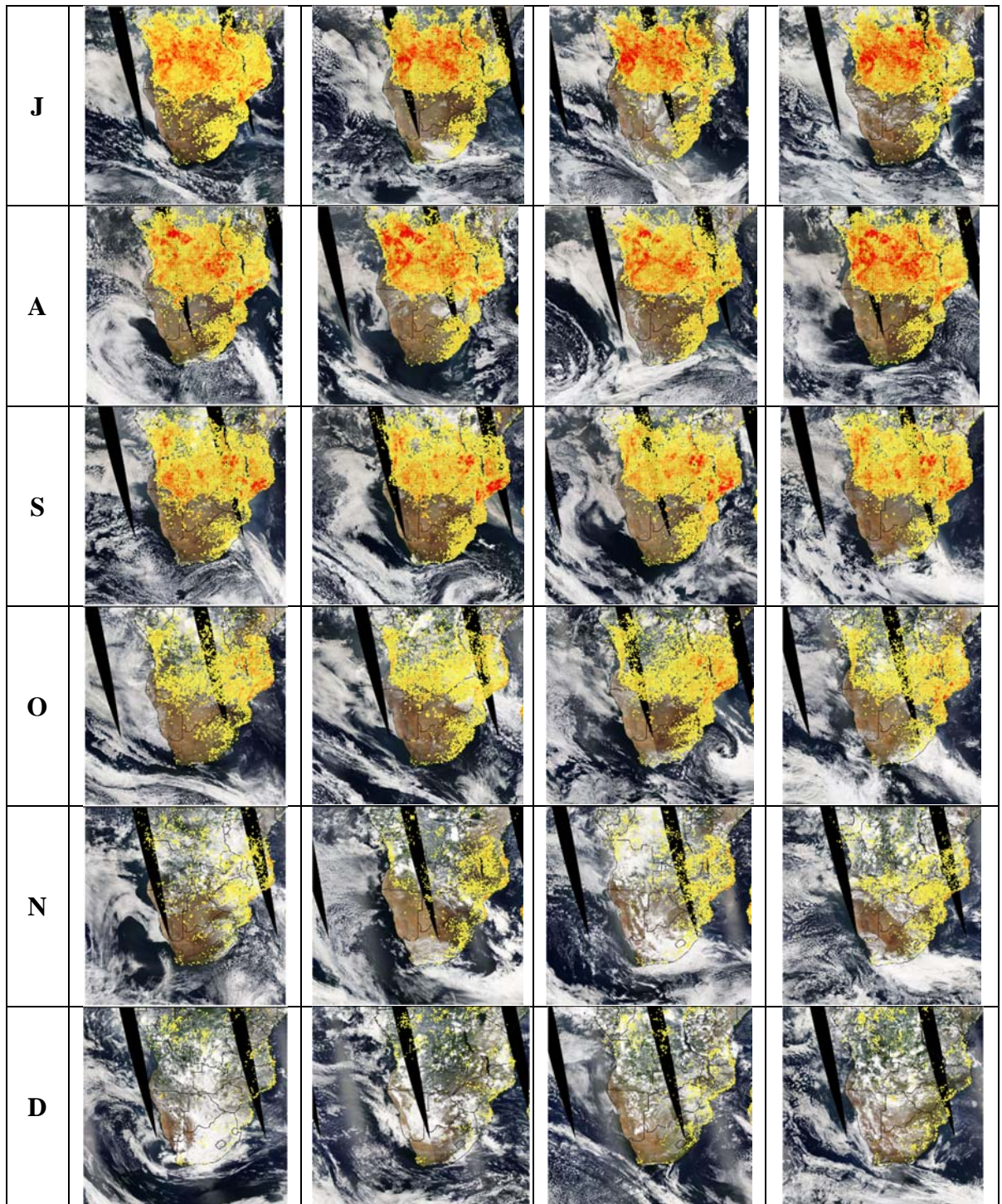
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 735 indicated over the first image  
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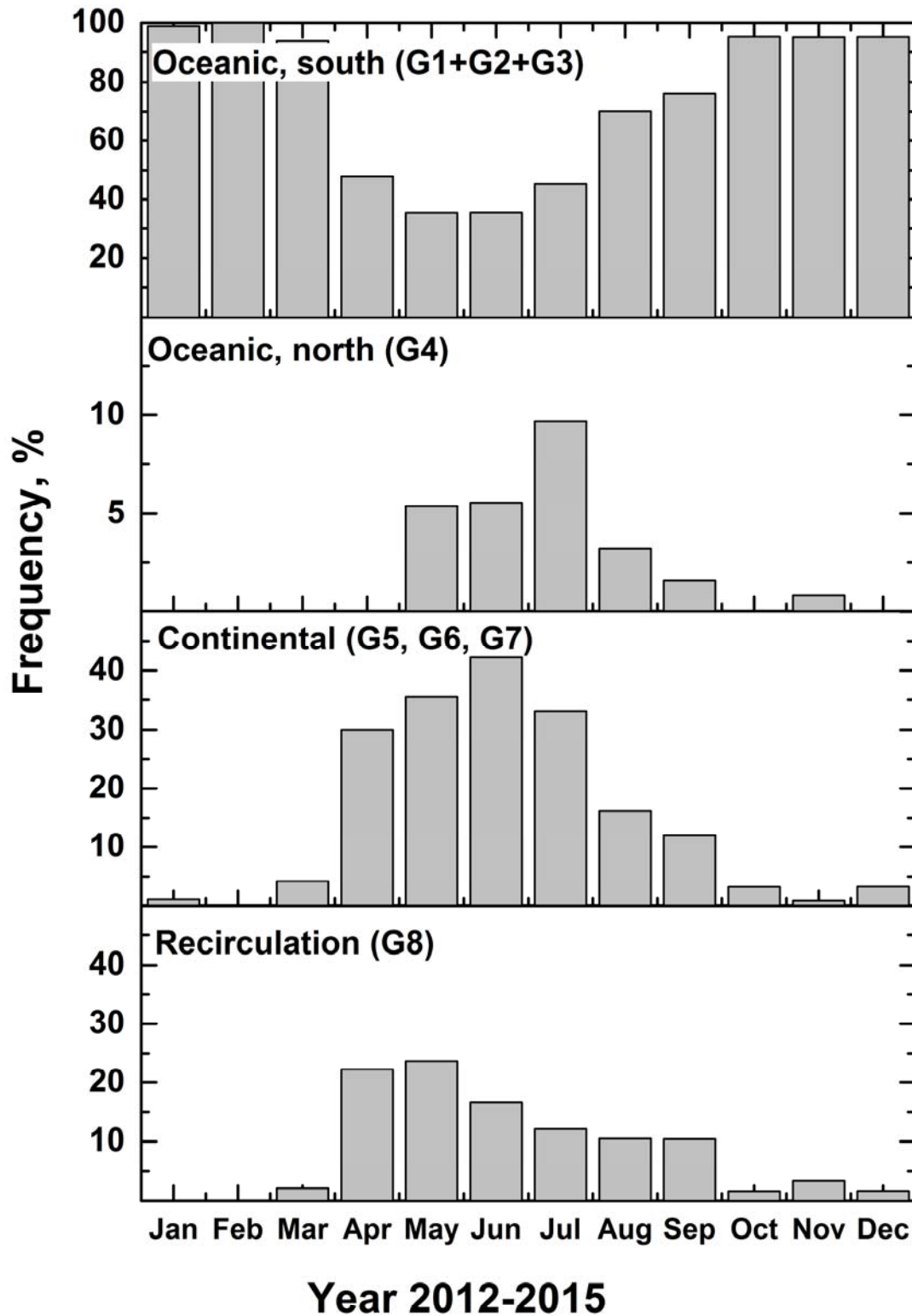




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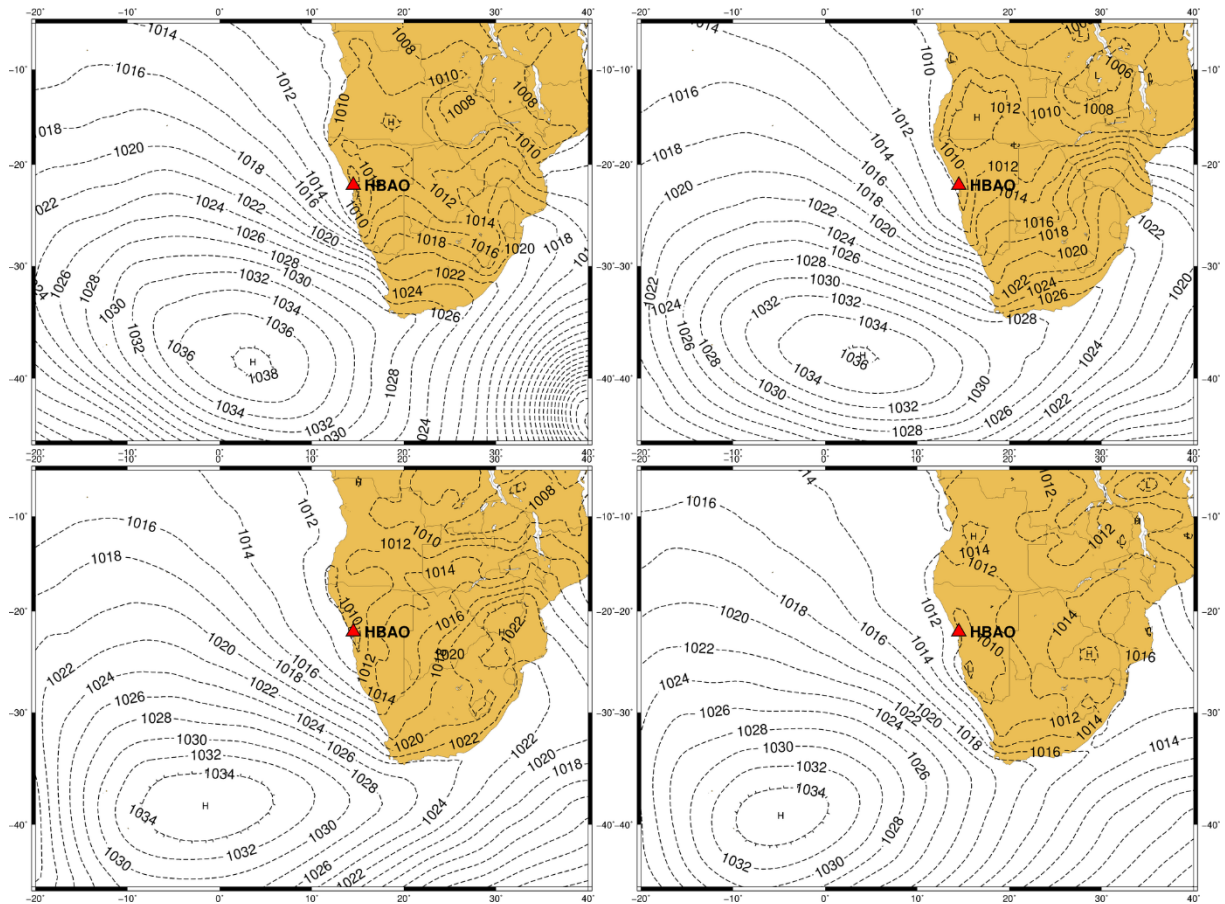
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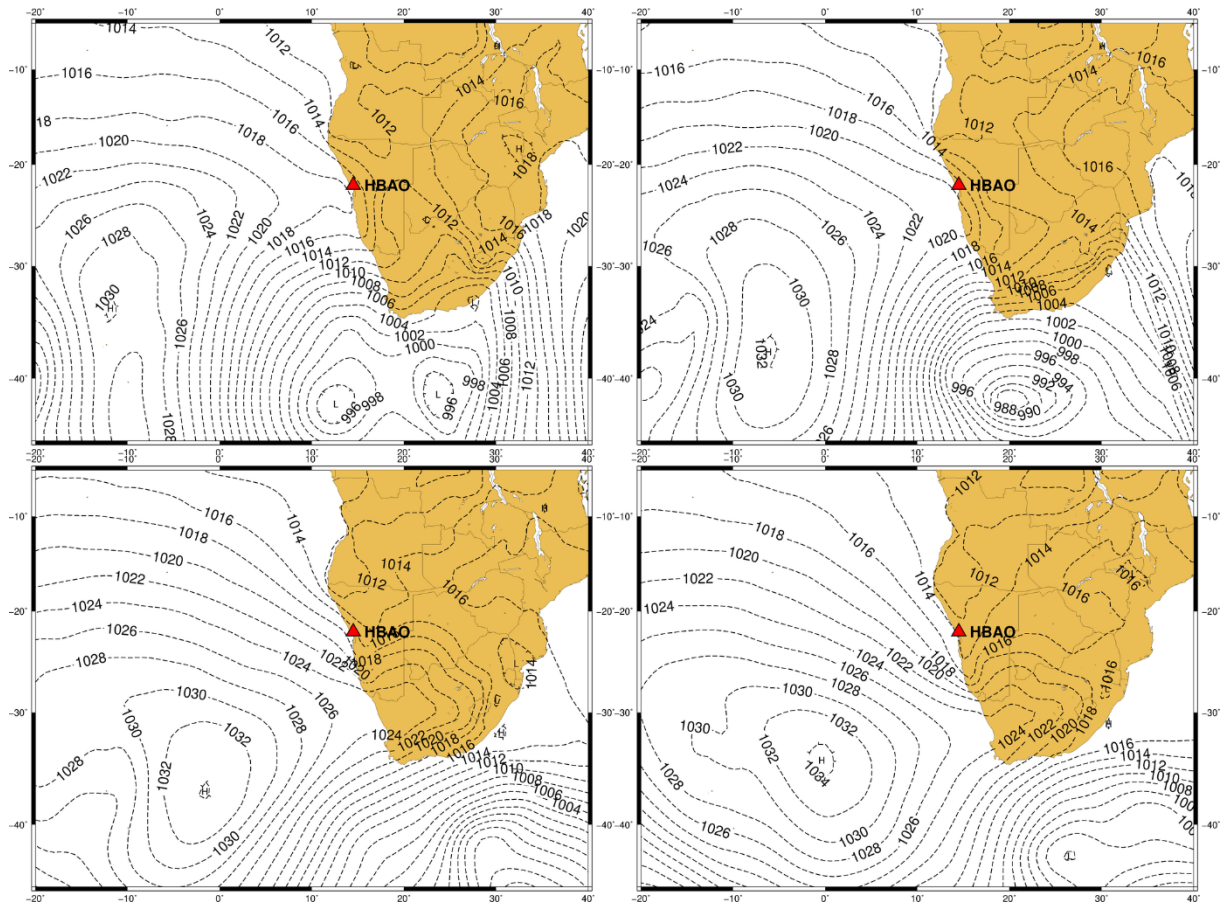
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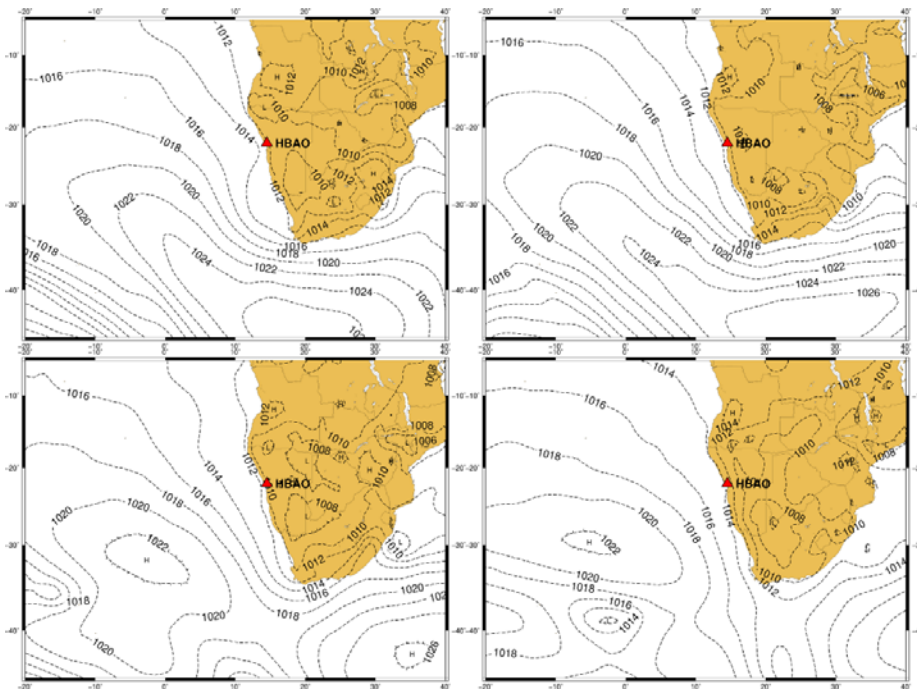
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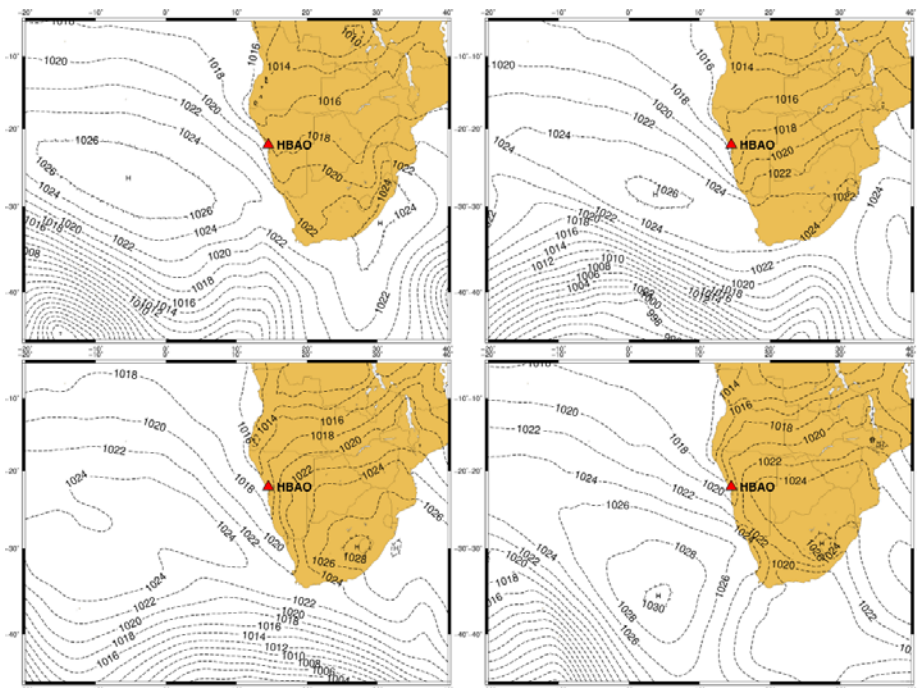
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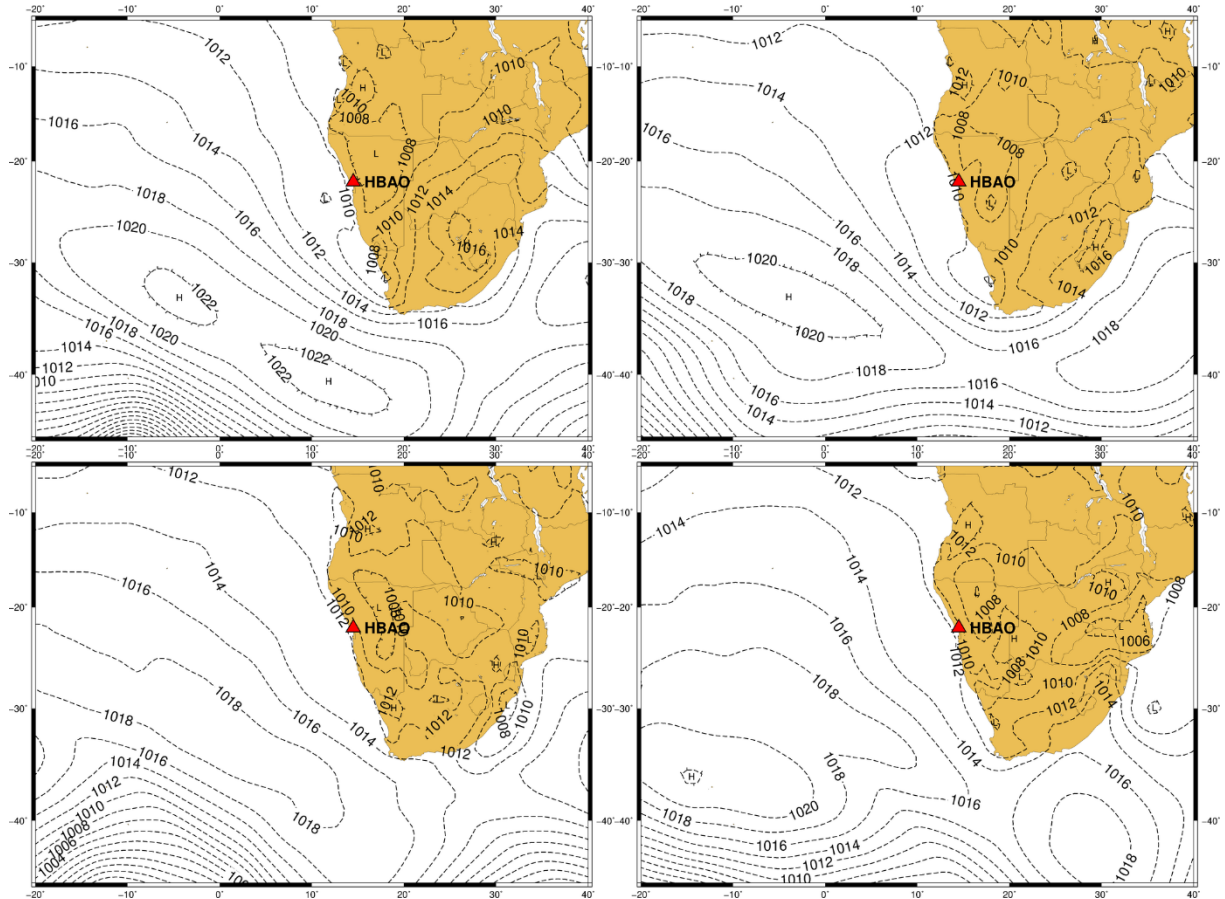


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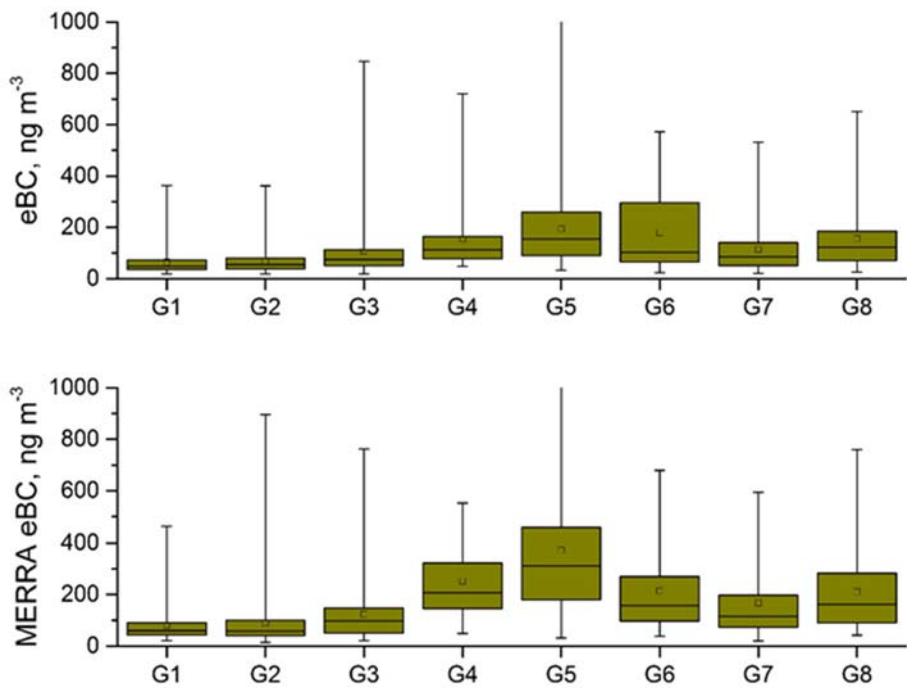
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