

1 **Precipitation-driven gamma radiation enhancement**
2 **over the Atlantic Ocean**

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7 **Key Points:**

- 8 • Precipitation-driven enhancements in gamma radiation are detected in the oceanic
9 environment.
10 • Gamma radiation enhancements are found in the open ocean at large distances
11 (+ 500 km) from the nearest coastline.
12 • Rain events do not produce enhancements in gamma radiation, even close to the
13 coast, for marine air masses with no recent contact with land.

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Abstract

Gamma radiation over the Atlantic Ocean was measured continuously from January to May 2020 by a NaI(Tl) detector installed on board the Portuguese navy’s ship NRP Sagres. Enhancements in the gamma radiation values are identified automatically by an algorithm for detection of anomalies in mean and variance as well as by visual inspection. The anomalies are typically +50% above the background level and relatively rare events ($\sim < 10\%$ of the days). All the detected anomalies are associated with simultaneous precipitation events, consistent with the wet deposition of scavenged radionuclides. The enhancements are detected in the open ocean even at large distances (+ 500 km) from the nearest coastline. Back trajectories reveal that half of these events are associated with air masses experiencing continental land influences, but the other half don’t display evidence of recent land contact. The enhancements in gamma radiation very far from land and with no evidence of continental fetch from back trajectories are difficult to explain as resulting only from radionuclides with a terrestrial source such as radon and its progeny. Further investigation and additional measurements are needed to improve understanding on the sources of ambient radioactivity in the open ocean and assess whether gamma radiation in the marine environment is influenced not only by radionuclides of terrestrial origin, but also cosmogenic radionuclides, like Beryllium-7, formed in the upper atmosphere but with the ability to be transported downward and serve as a tracer of the aerosols to which it attaches.

Radioactive elements such as the noble gas radon and those produced by its radioactive decay are naturally present in the environment and used as tracers of atmospheric transport and composition. In particular the noble gas radon, being inert and of predominantly terrestrial origin, is used to identify pristine marine air masses with no land contamination. Precipitation over land typically brings radon from the atmosphere to the surface, enhancing gamma radiation on the ground, but over such enhancements have not been identified before nor expected over the ocean due the low amount of radon typical of marine air masses. Here we report, for the first time, gamma radiation enhancements associated with precipitation in the oceanic environment, using measurements performed over the Atlantic ocean in a campaign onboard the Portuguese navy sip NRP Sagres.

1 Introduction

Gamma radiation is well known to exhibit significant enhancements associated with precipitation events (e.g. Fujinami (1996); Yakovleva et al. (2016); Bossew et al. (2017); Melintescu et al. (2018)). The increase in gamma radiation results mainly from the wet deposition of the progeny of Rn-222 (radioactive half-life = 3.82 days), mainly Pb-214 and Bi-214 (e.g. Livesay et al. (2014); Bottardi et al. (2020); Zelinskiy et al. (2021)). The gamma radiation peaks typically exhibit a short time rise and a longer decrease time resulting from the direct deposition of Pb-214 and Bi-214 on the ground and subsequent decay, with gamma radiation remaining above background values for several half-lives, about 3-4 hours (Fujitaka et al., 1992; Greenfield et al., 2008; Livesay et al., 2014; Reuveni et al., 2017). The concentration of radon progeny in precipitation is not correlated with the concentration of radon progeny in air near the surface (Fujinami, 1996), suggesting that the scavenging of radionuclides to the ground is dominated by processes within the clouds - nucleation scavenging and interstitial aerosol collection by cloud or rain droplets - rather than by processes below the cloud base (e.g. Takeuchi and Katase (1982); Paatero and Hatakka (1999)). The increase in gamma radiation associated with precipitation depends on the history of the corresponding contributing air mass (Paatero, 2000; Inomata et al., 2007; Mercier et al., 2009; S. Barbosa et al., 2017) but no clear association has been found between precipitation (intensity, amount and duration), and the resulting enhancement in gamma radiation (Fujinami, 1996; Burnett et al., 2010; Cortes et al., 2001; Greenfield et al., 2003; Datar et al., 2020). The connection between the temporal variability

65 of gamma radiation and precipitation is not straightforward as a result of the complex
66 interplay of factors such as the amount and intensity of precipitation, the cloud's thick-
67 ness and base height, and the atmospheric concentration of sub-micron aerosols, all in-
68 fluencing the scavenging of radon progeny (e.g. S. Barbosa et al. (2017)).

69 Although gamma radiation peaks driven by precipitation have been studied in num-
70 erous and varied settings, here we report, for the first time, gamma radiation enhance-
71 ments associated with precipitation in the oceanic environment. Measurements of total
72 gamma radiation have been performed in open ocean over the North and South Atlantic
73 in the framework of project SAIL - Space-Atmosphere-Ocean Interactions in the marine
74 boundary Layer (S. Barbosa, Dias, et al., 2022), in a field campaign inspired by the Carnegie
75 expedition and its contribution to understanding the global atmospheric electric field (Harrison,
76 2013, 2020).

77 Over the ocean radon exhalation from the surface is negligible. The total oceanic
78 contribution to radon in the global atmosphere is only about 2% of all radon exhaled from
79 continents (Wilkening & Clements, 1975). Using a gas transfer model, Schery and Huang
80 (2004) derived an oceanic radon flux of $0.00182 \text{ atom cm}^{-2} \text{ s}^{-1}$, with the model indi-
81 cating strong spatial variability associated to its dependence on surface wind speed. Emis-
82 sion of radon from the ocean was taken by B. Zhang et al. (2021) as $0.005 \text{ atom cm}^{-2}$
83 s^{-1} , 200 times less than land emissions. The negligible oceanic contribution enables radon
84 to be used as an unambiguous indicator of recent terrestrial influence on an air mass (e.g.
85 Wilkening (1981); Balkanski et al. (1992)) and many studies have used radon to iden-
86 tify continental fetch areas and long-range transport from terrestrial source regions (e.g.
87 Polian et al. (1986); Zahorowski et al. (2005); Chambers et al. (2013, 2018); Jun et al.
88 (2022)).

89 Unlike radon, which is inert and neutral, radon progeny are mostly positively charged
90 and react with water vapor and trace gases, forming clusters of small particles that are
91 quickly and irreversibly attached to existing aerosols in the atmosphere (Whittlestone,
92 1990; Postendorfer et al., 1994; Bigg, 1996; Porstendörfer, 2001; Elsässer et al., 2011).
93 Therefore the fate of gamma-emitting radon progeny, after their formation by radioac-
94 tive decay, is closely linked to that of aerosols, particularly accumulation mode aerosol
95 particles with a diameter of a few hundred nanometers (Paatero et al., 2017). Observa-
96 tions of aerosol concentration over the ocean are limited, but deposition of aerosols to
97 the surface ocean, particularly the open ocean away from continental land masses, is an
98 important phenomena affecting marine biogeochemical cycles (e.g. Wei et al. (2022)).
99 Radioactive aerosols of radon progeny are deposited onto the Earth's surface primarily
100 by precipitation as accumulation-mode aerosols are too small for gravitational settling
101 and too large to be deposited by Brownian motion (F. Zhang et al., 2021).

102 In a marine setting gamma radiation variability mainly reflects atmospheric rather
103 than surface contributions. In terms of surface sources, gamma emission from the ocean
104 by radon degassing is negligible. The contribution from terrestrial sources containing ura-
105 nium and thorium and their decay series, which is substantial over land, is reduced over
106 the ocean. Gamma radiation from radionuclides in ocean sediments is attenuated by wa-
107 ter and doesn't reach the surface. In sea water potassium (K-40 isotope) is the domi-
108 nant gamma-emitting radionuclide, but it has a fairly uniform geographic distribution
109 (Solomon, 1988). In terms of atmospheric contributions, these include secondary cosmic
110 radiation, gamma rays resulting from the interaction of cosmic rays with gas molecules
111 in the atmosphere (e.g. Wissmann et al. (2005); Mertens (2016)), and airborne radionu-
112 clides. Airborne gamma-emitting elements include radon progeny (short-lived Pb-214,
113 Bi-214 and long-lived Pb-210) and cosmogenic radionuclides such as Be-7 (e.g. Bossew
114 et al. (2017); European Commission (2019)).

115 In the present study we document enhancements in gamma radiation over the At-
116 lantic ocean from high-resolution gamma radiation measurements. The data are described
117 in section 2, the analysis is detailed in section 3 and concluding remarks are provided
118 in section 4.

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2 Data

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Data considered in this study consist of gamma radiation (section 2.1) and meteorological measurements (section 2.2) performed over the Atlantic ocean from January to May 2020 on board the sail ship NRP Sagres. Figure 1 shows the map of the ship's trajectory since its departure from Lisboa in January 5th 2020. The trip was initially planned to last for 371 days, but was interrupted due to the COVID-19 pandemic and subsequent restrictions in port activities. On March 25th the ship arrived to Cape Town for refueling and supplies, departing the same day back to Portugal, instead of resuming the trip into the Indian Ocean as originally planned. The ship arrived to Lisboa on May 10th, after a stop for repairs at the port of Praia, Cape Verde. Overall data completion is $> 95\%$, with two short periods of data loss due to issues in the onboard computer and storage systems, which occurred on March 8th and 9th (during the trip from Buenos Aires to Cape Town) and then from 4 to 6 April, in the leg from Cape Town to Lisboa.

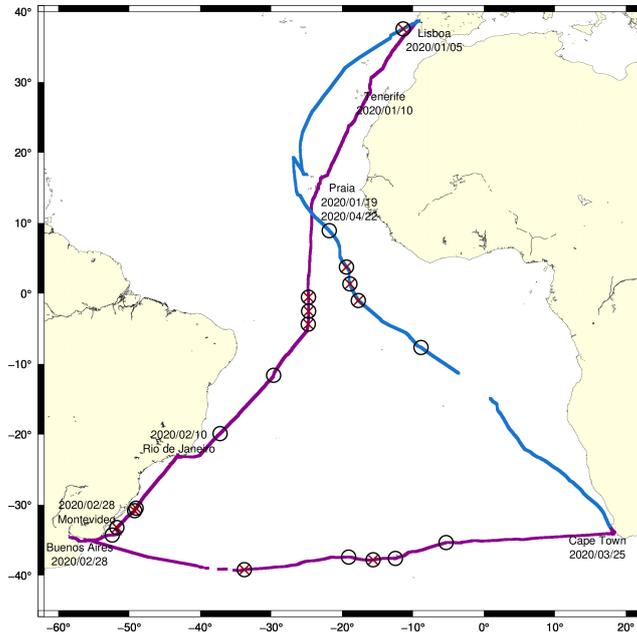


Figure 1. Map of the trajectory of NRP Sagres ship. The data points represented by light blue correspond to the Lisboa - South Africa leg of the trip, and darker blue represents the return trip from South Africa to Lisboa. The symbols ○ mark the location of the rain events listed in Table S1 and symbols × represent the location of the gamma anomalies listed in table 1. Blanks denote points with no available data due to computer issues ($< 5\%$ of the total data collected).

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2.1 Gamma radiation data

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Gamma measurements are performed with a $3'' \times 3''$ (76×76 mm) NaI(Tl) cylindrical scintillator (Scionix, the Netherlands) equipped with an electronic total count single channel analyzer for acquiring total counts of gamma radiation in the 475 keV to 3 MeV energy range. The selection of this energy range enables the reduction of Compton background in the 50–475 keV low-energy range, improving the sensitivity of short-lived radon progeny measurements (Zafir et al., 2011). The NaI(Tl) scintillator is encased in a water-proof container designed for underwater measurements, in order to pro-

141 tect the instrument from harsh marine conditions. The sensor is installed on the mizzen
 142 mast of the ship, at a height of ~ 20 m, in an upright position and pointing upwards.
 143 Counts are acquired at a sampling rate of 1-second and further aggregated into counts
 144 per minute. Further details on data management and pre-processing are described in the
 145 SAIL project’s data management plan (S. Barbosa & Karimova, 2021).

146 The 1 minute time series of gamma radiation counts is presented in Figure 2. Ex-
 147 cept for the evident ocean-land contrast, the temporal variation of gamma radiation counts
 148 is small, being more prominent in the first month of the series and very stable afterwards.
 149 The long-term component of gamma radiation variability is estimated by robust local
 150 regression (Cleveland et al., 1992) and represented by the colored solid line in Figure 2.
 151 The measurements performed over land during the stops of the ship along its journey,
 152 represented in gray in Figure 2 (top), are not further considered, as this work focus only
 153 on the observations of gamma radiation over the ocean. Thus the gamma radiation time
 154 series considered hereafter, displayed in Figure 2 (bottom), consists of the 1-minute gamma
 155 radiation counts measured exclusively in the marine environment (126 days in total).

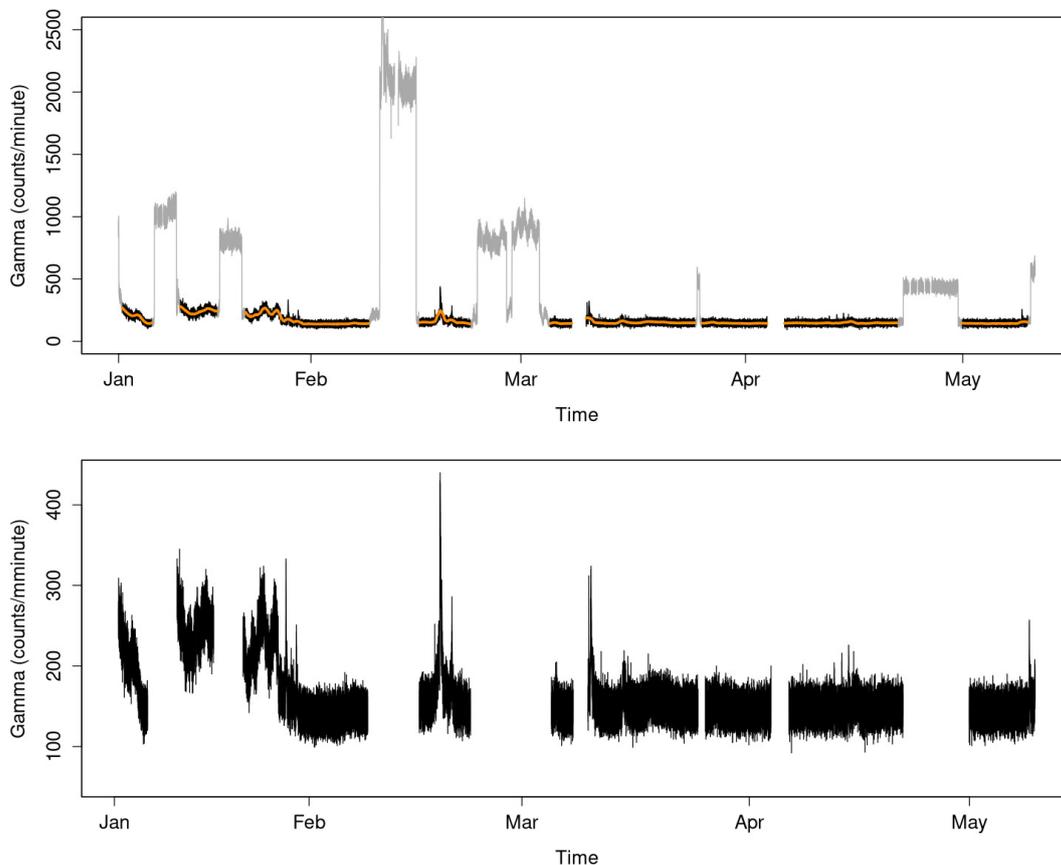


Figure 2. Time series of gamma radiation data. Top: complete 1-minute series with land measurements represented in gray and long-term variability by the solid colored line. Bottom: time series of marine-only 1-minute gamma radiation counts.

156 2.2 Meteorological data

157 Two distinct types of meteorological data are available from the SAIL campaign:
 158 automatic data collected by sensors, with no need of human intervention, and data col-
 159 lected by human observers. The meteorological optical range is measured every 1-minute

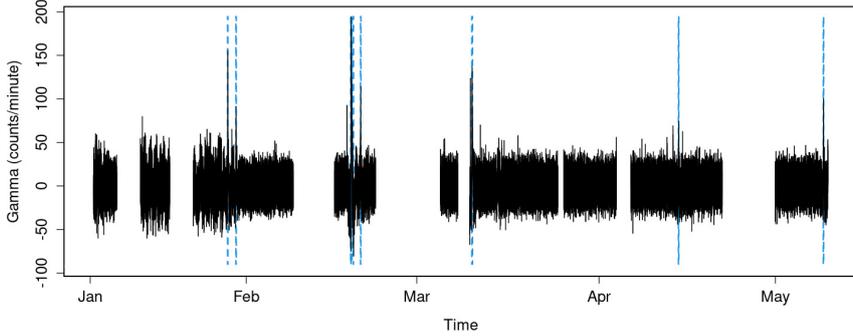


Figure 3. Detrended time series of gamma radiation. The anomalies identified by the CAPA algorithm are represented by the vertical dashed lines.

160 by a visibility sensor SWS050 (Biral, UK) providing measurements in the range from 10m
 161 to 40 km. The visibility sensor is located at the same height and on the same mast as
 162 the gamma radiation instrument. Rain, and basic meteorological parameters such as at-
 163 mospheric pressure, temperature and wind, are collected in a non-automatic way by the
 164 ship’s crew every 1-hour as part of the navy’s operational routine during navigation (no
 165 meteorological information is available when the ship is docked). Rainfall events are recorded
 166 in a qualitative way (drizzle < light < moderate). The geographic location of rain events
 167 is shown as \circ in Figure 1. Table S1 summarizes the available information in terms of
 168 rain occurrences during the whole trip. In general rain was not a frequent event, as it
 169 is registered in only 16 days out of a total of 126. Times were originally recorded as lo-
 170 cal time but are presented as coordinated universal time (UTC), as for all the other data.
 171 Rain registered at a given hour corresponds to rain observed within the previous hour.

172 3 Analysis

173 3.1 Detection of gamma radiation anomalies

174 For the detection of anomalies in the marine gamma radiation time series (Fig. 2,
 175 bottom), two complementary distinct approaches are used: an automatic method and
 176 visual inspection of the time series. The automatic detection of anomalies is performed
 177 using the Collective And Point Anomaly (CAPA) algorithm (Fisch et al., 2022). The out-
 178 comes of the algorithm are very much dependent on the pre-processing of the time se-
 179 ries in terms its standardization and handling of missing values. This is particular crit-
 180 ical in this case due to the numerous gaps in the time series. Thus for the application
 181 of the CAPA procedure the following pre-processing steps are taken: i) the long-term
 182 variability signal (represented by the solid line in Fig. 2 top) is subtracted from the se-
 183 ries for stabilization of the mean; and ii) the gaps are filled by replacing the missing
 184 values by values resulting from a normal distribution with the same mean and variance as
 185 the gamma radiation time series. The CAPA algorithm is then applied to the pre-processed
 186 time series using a penalty for control of false positives of $2 \times \frac{1+\phi}{1-\phi} \log(n)$, where ϕ is set
 187 as 0.9 and n is the length of the time series. The results are displayed in Figure 3.
 188 In a conservative approach (mainly determined by the penalty value for control of false
 189 positives), a total of 8 anomalies are detected. Visual inspection confirms these, and fur-
 190 ther identifies 4 additional candidate anomalies in gamma radiation, summarized in Ta-
 191 ble 1. The geographic location of these 12 anomalies is displayed in Figure 1.

Table 1. Anomalies identified in the marine gamma radiation observations by visual inspection and by using the CAPA algorithm. It is also indicated whether these periods identified as anomalous correspond to rain events or anomalies in visibility.

date	time (UTC)	Visual detection	CAPA algorithm	Rain	Visibility
2020-01-28	19:00-21:00	✓	✓	✓	✓
2020-01-29	13:00-14:00	✓	-	✓	✓
2020-01-30	05:00-07:00	✓	✓	✓	✓
2020-02-18	19:00-24:00	✓	✓	✓	✓
2020-02-19	01:00-02:00	✓	✓	-	✓
2020-02-20	10:00-12:00	✓	✓	-	✓
2020-03-10	08:00-16:00	✓	✓	✓	✓
2020-03-15	10:00-11:00	✓	-	-	✓
2020-04-12	14:00-16:00	✓	-	✓	✓
2020-04-13	14:30-15:30	✓	-	-	✓
2020-04-14	13:00-14:00	✓	✓	✓	✓
2020-05-09	04:00-06:00	✓	✓	✓	✓

Table 2. Contingency table for the number of occurrences (in days) of rain and gamma radiation anomalies.

	number of days rain	number of days no rain	
gamma anomaly	8	4	12
no gamma anomaly	8	106	114
	16	110	126

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3.2 Characteristics of marine gamma anomalies

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Table 2 summarizes the occurrence of anomalies in the gamma radiation time series as a function of the rainfall information. From a total of 126 days with gamma radiation measurements over the ocean, gamma anomalies are identified in only 12 days (< 10%). Most of these anomalies ($\sim 65\%$) are associated with the occurrence of rain according to the available meteorological information from human observers. They are also associated with concurrent anomalies in the meteorological optical range from the visibility sensor, as illustrated in Figure 4. Only 4 gamma radiation anomalies occur in days for which rain was not registered by human observers. And in all these 4 cases the anomalies in gamma radiation are associated with simultaneous sharp drops in visibility, as shown in Figure 5. Thus it seems likely that also these gamma radiation anomalies are driven by precipitation which apparently failed to be registered by the human observers.

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Although all enhancements in gamma radiation are associated with the occurrence of precipitation, the reverse is not true, i.e. the occurrence of precipitation is not necessarily associated with an anomaly in gamma radiation. For a total of 16 days with registered rain events, half do not have a corresponding anomaly in the gamma radiation counts. These cases are detailed in Figures 6 and 7. Comparison of the visibility measurements with the meteorological information in Table S1 shows strong consistency between human-recorded and instrumental information. Only in one case (16th April 2020 - Figure 7) the visibility data does not point to the occurrence of rain, in disagreement with the qualitative information of early morning drizzle. In all the remaining cases vis-

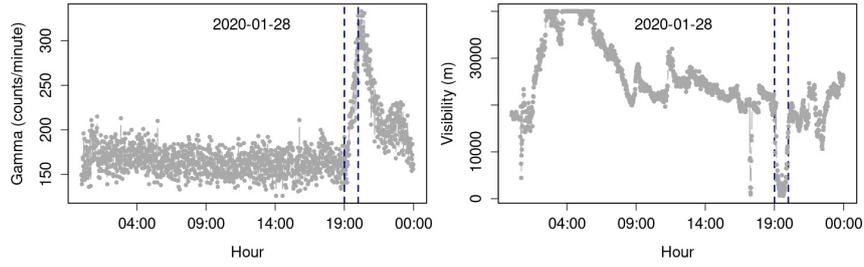


Figure 4. Detail (28th January 2020) of 1-minute time series of gamma radiation counts (left) and visibility (right). The vertical dashed lines represent the period of occurrence of moderate rain as indicated in the available meteorological information.

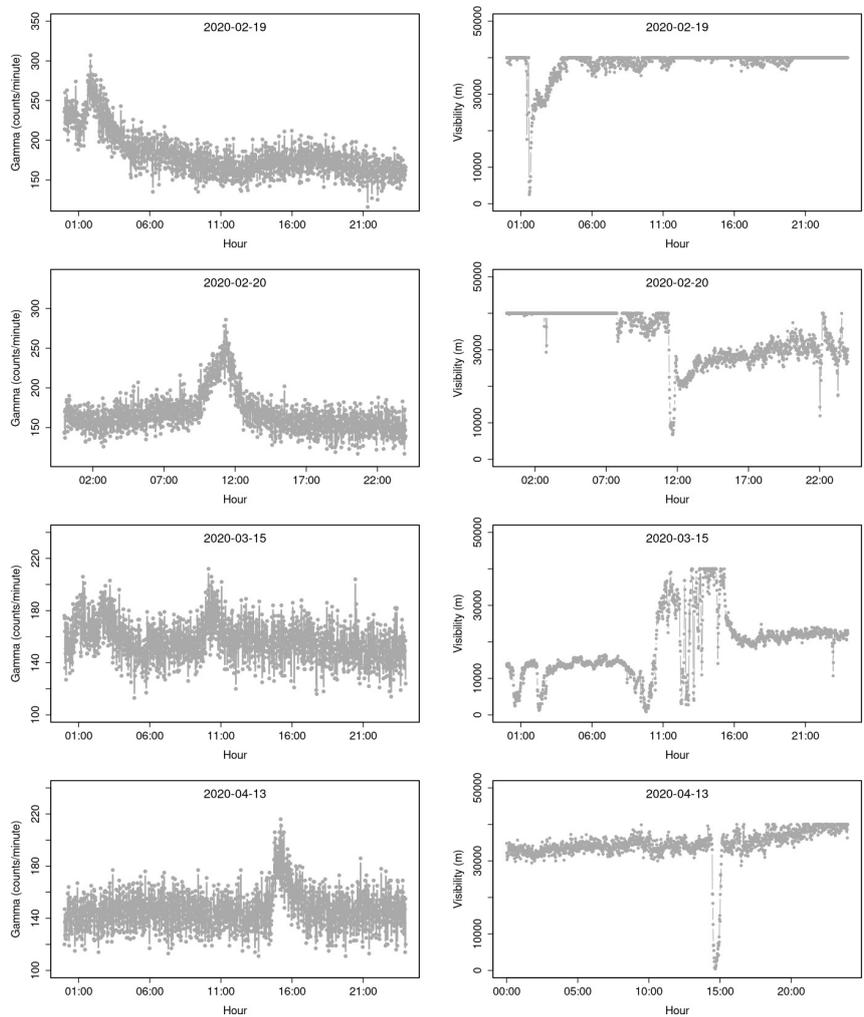


Figure 5. Detail of 1-minute time series of gamma radiation counts (left) and visibility (right) for the days in which an anomaly is identified in gamma radiation but rain is not registered in the navy’s meteorological observations.

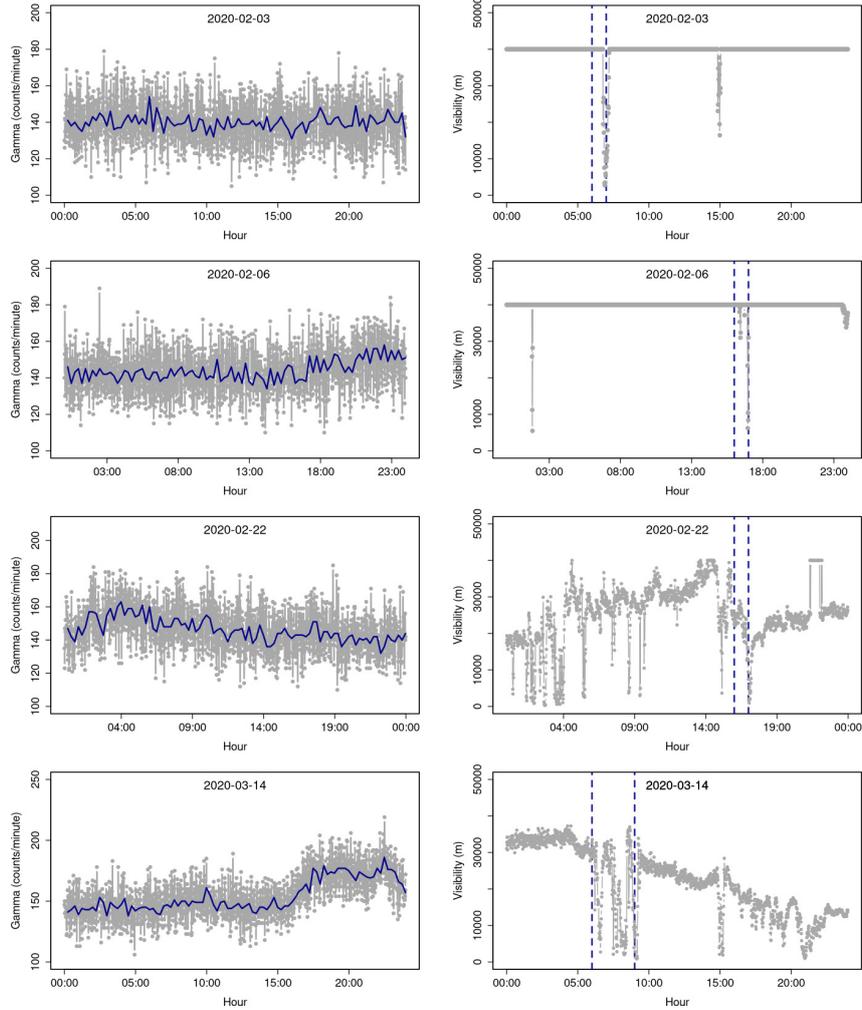


Figure 6. Detail of 1-minute time series of gamma radiation counts (left) and visibility (right) for the days with occurrence of precipitation but no gamma anomalies. The solid (blue) line represents the 15-minute running median of gamma radiation counts. The vertical dashed lines represent the period of occurrence of rain from the available meteorological information.

214 visibility measurements are very consistent with the qualitative rain data information avail-
 215 able. Thus the absence of gamma anomalies (or in two cases - 2020-03/18 and 2020-04-
 216 08 - only very small increases barely detectable within the noise level) is not related to
 217 eventual errors in the qualitative rain information.

218 Table 3 shows the % enhancement in gamma radiation and the corresponding distance
 219 to the nearest coastline for all days with an anomaly in gamma radiation and/or
 220 occurrence of rain. The % enhancement is obtained for each day in which a gamma anomaly
 221 was identified by computing the difference of the maximum gamma value relative to the
 222 average background value of that day. The distance to the nearest coastline is computed
 223 using the Generic Mapping Tools (GMT) software (Wessel et al., 2019) using its low res-
 224 olution coastline (Wessel & Smith, 1996). Inspection of Table 3, Figure S1, displaying
 225 the % increase in gamma radiation as a function of the distance to the nearest coastline
 226 and rain characteristics, (and also of the map in Figure 1) doesn't reveal any clear as-
 227 sociation between gamma radiation anomalies and the type of precipitation as qualita-

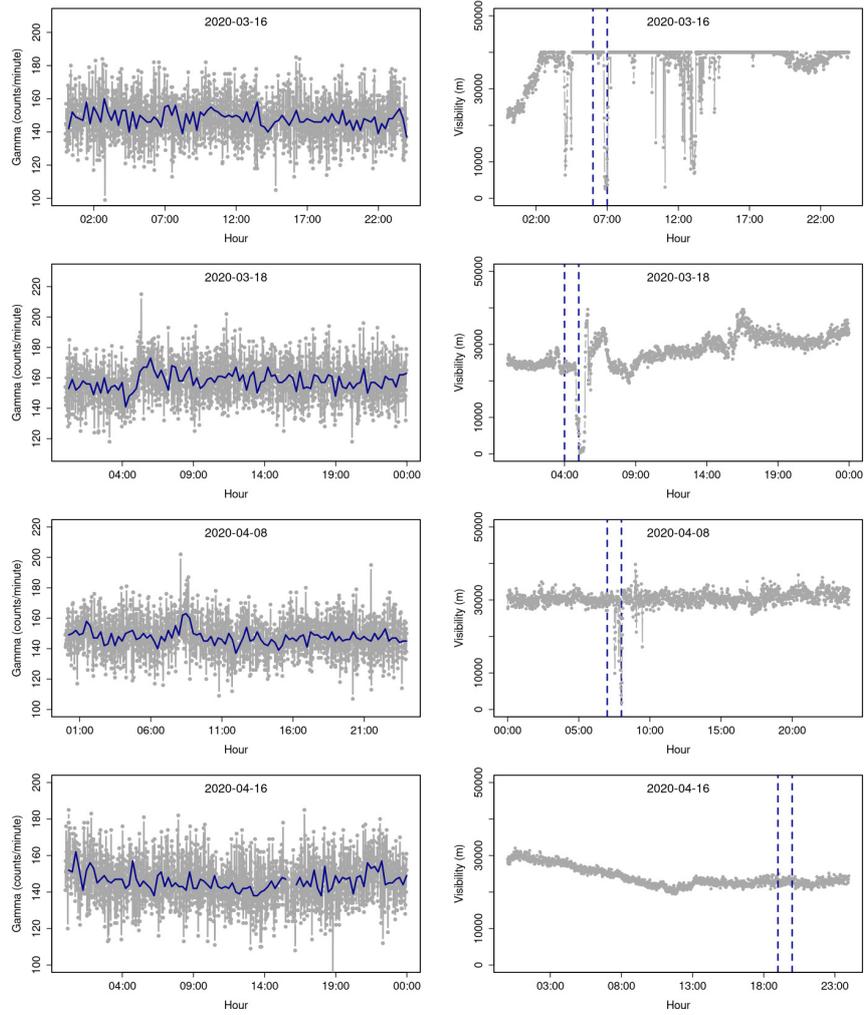


Figure 7. same as in Figure 6.

228 tively recorded by human observers. Furthermore, no consistent association was observed
 229 between the enhancement in gamma radiation and the distance to the nearest landmass.

Table 3. Approximate distance to the nearest coastline for all the days with an anomaly in gamma radiation and/or occurrence of rain. (1) denotes days in which rain is inferred from visibility measurements and (2) rain occurrence suspect (not confirmed by visibility data).

date	rain	increase in gamma (%)	distance to land (km)
2020-01-28	moderate	99	927
2020-01-29	drizzle	33	866
2020-01-30	drizzle	70	849
2020-02-03	drizzle	-	677
2020-02-06	drizzle	-	272
2020-02-18	drizzle	142	118
2020-02-19	(1)	78	112
2020-02-20	(1)	79	81
2020-02-22	drizzle	-	105
2020-03-10	moderate	95	1666
2020-03-14	light/moderate	-	564
2020-03-15	(1)	36	263
2020-03-16	light	-	35
2020-03-18	drizzle	-	649
2020-04-08	light	-	600
2020-04-12	moderate	40	847
2020-04-13	(1)	49	948
2020-04-14	drizzle	54	820
2020-04-16	(2)	-	639
2020-05-09	drizzle	73	213

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3.3 Back trajectories

231 Distance to the coast alone is not an unambiguous criterium to assess continental
 232 influences on the marine atmosphere. Air mass back trajectories can be a powerful tool
 233 for interpreting fetch behavior, particularly in the absence of local meteorological mea-
 234 surements (e.g. Chambers et al. (2013)). Back trajectories were computed with the HYS-
 235 PLIT transport and dispersion model (Stein et al., 2015), version 5.2.2, using meteorological
 236 information from the Global Data Assimilation System with 1 degree resolution
 237 (GDAS1). The 10-day back trajectories were computed at two distinct heights (500m
 238 and 2000m) for all the 19 rain events listed in Table 4 (excluding only the 16th April event
 239 for which the occurrence of rain is questionable). These heights were chosen to be rep-
 240 resentative of air masses within, and outside of, the marine boundary layer, respectively.

241 The back trajectories results are displayed in Figures S2 to S3 and Figure 8 cor-
 242 responding to 3 distinct cases: i) back trajectories showing no evidence of recent land
 243 contact, and for which rainfall does not produce a gamma anomaly (Figure S2); ii) back
 244 trajectories showing clear or at least some indication of continental fetch, and for which
 245 gamma anomalies are identified (Figure S3); and iii) back trajectories suggesting no re-
 246 cent contact of the air masses with land, but for which rainfall produces rain anomalies
 247 (Figure 8). The remaining rainfall event on 2020-03-18 corresponds to a very small gamma
 248 anomaly and an air mass with some evidence of land contact. The results are summa-
 249 rized in Table 4. The back trajectories for the rainfall events not associated with a peak
 250 in gamma radiation (or a very small anomaly, in the case of the March 18th event), sug-
 251 gest in all those 7 cases no contact with land or at least for the February 22th, March

Table 4. Contingency table for gamma radiation anomalies and land influences derived from 10-days back trajectories.

	land contact	no land contact	
gamma anomaly	6	6	12
no gamma anomaly	0	7	7
	6	13	19

252 16th and 18th cases no recent land influence (Figure S2). In the case of the 12 rain events
 253 with corresponding enhancement in gamma radiation, half of them seem to correspond
 254 to air masses with continental influences (Figure S3), while the other half doesn't dis-
 255 play evidence of recent land contact (Figure 8). The smallest enhancements in gamma
 256 radiation correspond to cases where back trajectories suggest no recent contact of air masses
 257 with land, and the largest enhancement corresponds to a location near land, with ev-
 258 ident terrestrial influence (Figure 9).

259 4 Discussion and conclusions

260 This work documents, for the first time, enhancements of gamma radiation over
 261 the ocean associated with the occurrence of precipitation. Most of these enhancements
 262 were observed in the southern hemisphere and at varying distances from land, from about
 263 100 km to more than 1500 km to the nearest shoreline.

264 All the enhancements identified in the marine gamma radiation time series are as-
 265 sociated with concurrent occurrence of rain (either explicitly registered by human ob-
 266 servation or inferred by visibility data). This fact is consistent with the wet deposition
 267 mechanism being the main driver of ground enhancements in gamma radiation.

268 As it is also the case for gamma radiation enhancements over land, a clear asso-
 269 ciation between the magnitude of the gamma anomaly and the amount and intensity of
 270 precipitation is not discernible in this study, although here the analysis is limited by the
 271 short length of the time series (5 months), and by the low temporal resolution (1 hour)
 272 and the qualitative nature of precipitation observations. Still the information from hu-
 273 man observation is in very good agreement with the meteorological optical range mea-
 274 sured by the visibility sensor, giving confidence to the use of both types of data.

275 No systematic relationship is observed between the enhancement in gamma radi-
 276 ation and the distance to land nor the air masses previous contact with land. An obvi-
 277 ous limitation to better quantification of such relationships is the small number of events
 278 under consideration (12), a longer time series would allow a more detailed assessment.

279 The back trajectories confirm no recent contact with land in all cases for which rain
 280 events do not produce an enhancement in gamma radiation (Figure S2). The oceanic fetch
 281 explains why enhancements in gamma radiation are not produced even for comparatively
 282 small distances to land (e.g. February 6th and 22nd events). In the 16th March case the
 283 distance to land is only 35 km, but the location is very far from continental land masses,
 284 near the Tristan da Cunha island in the South Atlantic. This confirms the expected low
 285 content of radon progeny in marine air masses away from continental sources.

286 The gamma radiation enhancements coincident with precipitation events occur, as
 287 expected, closer to land (February 18th, 19th 20th events), corresponding to air masses
 288 crossing continental regions, or in open ocean, in the tropical Atlantic region, with air
 289 masses back trajectories (Figure ??) suggesting the possibility of continental influences
 290 (particularly for the January 29th and 30th cases, not so clear for the 13th April case).

291 However, gamma radiation anomalies associated with precipitation events are also
 292 observed in the open ocean, very far from the coast, and for air masses for which back
 293 trajectories don't show evidence of recent contact with land (Figure 8). While in some

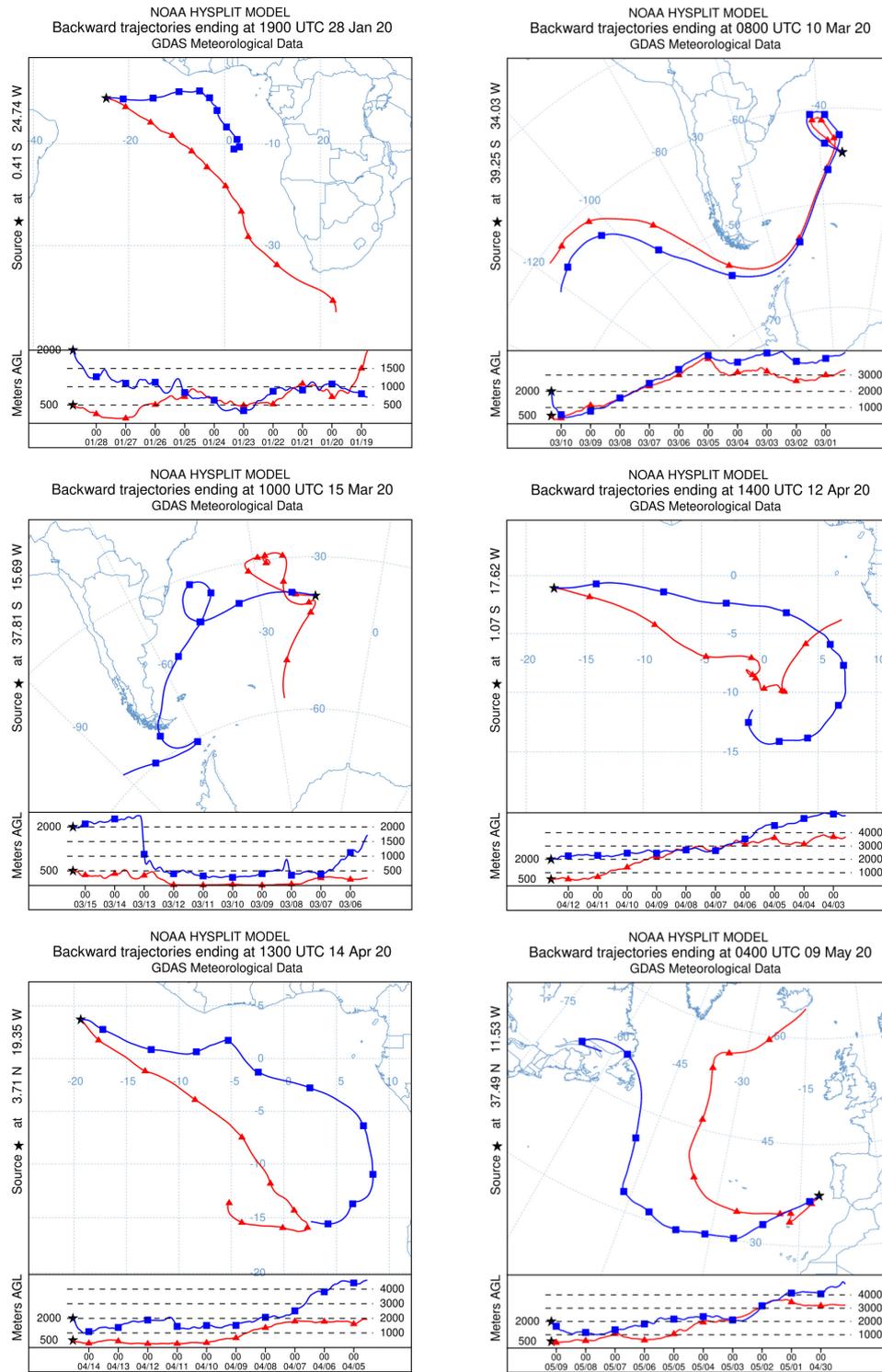


Figure 8. Examples of 10-days back trajectories suggestive of no recent contact with land for the case of rain events with corresponding gamma anomaly.

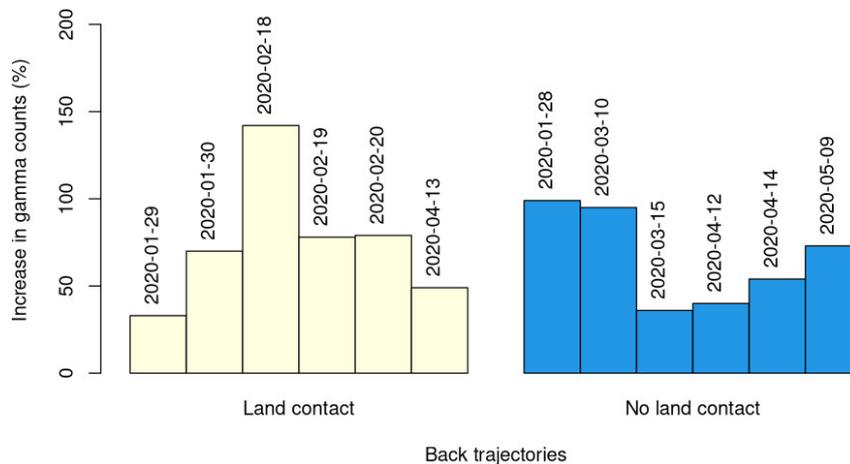


Figure 9. Barplots of the magnitude of the gamma anomalies associated with precipitation for the cases of air masses with recent land contact (left) and no land contact in the previous 10-days (right).

294 cases (January 28th and March 15th events) limitations in back trajectories can be the
 295 culprit - arguably the 28th January trajectory, while not crossing land, is not as differ-
 296 ent from the 29th and 30th January trajectories - the results show significant enhance-
 297 ments in gamma radiation very far from land and with no evidence of continental fetch
 298 from back trajectories results (March 10th, April 12th, April 14th and May 9th). These
 299 cases are difficult to explain as resulting only from radionuclides with a predominantly
 300 terrestrial source, such as radon and its progeny.

301 A further potential contribution to these enhancements in gamma radiation observed
 302 in open ocean and with no evidence of continental fetch is the gamma-emitting radionu-
 303 clide Beryllium-7 (Be-7), produced in the Earth's upper atmosphere by cosmic radia-
 304 tion through the spallation of nitrogen and oxygen (Lal, 1967). It has a half-life of \sim
 305 53 days, emitting gamma radiation with energy of \sim 477.6 keV (Tilley et al., 2002). Af-
 306 ter its formation Be-7 readily becomes associated with aerosols in the sub-micron size
 307 range (e.g (Winkler et al., 1998; Ioannidou et al., 2005; Elsässer et al., 2011)) and is then
 308 subject to complex horizontal and vertical atmospheric transport processes (Kaste et al.,
 309 2002). Precipitation scavenging is the dominant (\sim 90%) process of removal of Be-7
 310 from the atmosphere (Kaste et al., 2002; Kusmierczyk-Michulec et al., 2015; Mohan et al., 2019)
 311 and low precipitation rates during drizzles are particularly efficient in scavenging Be-7
 312 by fine droplets (Ioannidou & Papastefanou, 2006).

313 The cases reported here of gamma radiation enhancements in the open ocean with
 314 no apparent continental influences (10th March, 12th and 14th April and May 5th events)
 315 correspond according to the back trajectories displayed in Figure 8 to descending air masses.
 316 This is consistent with the expectation that concentrations of cosmogenic radionuclides
 317 such as Be-7 should increase due to the influx of air from the upper atmosphere enriched
 318 in Be-7 radionuclides (Doering & Saey, 2014).

319 The enhancements in total gamma radiation documented in the present study can't
 320 be unequivocally attributed to a specific radionuclide, as the measurements are of total
 321 gamma radiation in an energy range (0.475-3 MeV), optimal for radon progeny mea-
 322 surements but also including gamma radiation emitted by Be-7. Thus whether only progeny
 323 from airborne radon gas, even if present in small amounts, or other contributions (sec-
 324 ondary cosmic radiation, Be-7 radionuclides formed in the upper atmosphere) are respon-
 325 sible for the identified gamma anomalies cannot be settled from the available data. Fur-
 326 ther measurements would be required, in particular spectral gamma observations which

327 would allow to ascertain which specific elements are contributing to the measured to-
328 tal gamma radiation. An energy-discriminating sensor (Aplin et al., 2017) was actually
329 installed on board NRP Sagres in the framework of the SAIL campaign, but unfortu-
330 nately mal-functioning of the instrument prevented acquisition of data during the field
331 campaign. In terms of additional measurements direct radon gas concentration obser-
332 vations would be also very helpful, though a detector sensitive enough to be able to mea-
333 sure very low radon concentrations typical of marine air would be necessary (Chambers
334 et al., 2018).

335 Although the results presented here raise questions that can't be answered with-
336 out further investigation and collection of new data - a challenging endeavor in a ma-
337 rine setting - the identification of gamma anomalies far from landmasses and apparently
338 not under influence of long-range transport conditions suggests the possibility that not
339 only radon progeny but also other radionuclides, in particular Be-7, can contribute to
340 the identified anomalies in marine gamma radiation. The fact that radon progeny (as
341 well as Be-7) attach rapidly to aerosols after formation, suggests that gamma radiation
342 measurements from pristine, least influenced by land airmasses, could then be used as
343 a proxy of aerosols in the marine environment.

344 The time series of marine gamma observations (Figure 2, bottom) exhibits larger
345 values and also higher variability in January compared with the observations after Febru-
346 ary. The coupling between mean and variance is typical of radon progeny time series (S. M. Bar-
347 bosa et al., 2007), but the fact that very dissimilar background values are observed even
348 at the same location (note the contrast in marine background values at Cape Verde for
349 the two distinct legs of the ship route, at the end of January and then at the end of April)
350 is significant. Possible explanations include synoptic conditions favoring continental fetch
351 during that period and thus increased radon gas concentration and/or seasonal variabil-
352 ity of aerosols and wind regime leading to an increase in radon progeny and eventually
353 Be-7 radionuclides. This requires further investigation and a more detailed assessment
354 which is out of scope of the present study focusing on enhancements in gamma radia-
355 tion associated with precipitation.

356 Further investigation and additional measurements (energy-discriminating gamma
357 radiation observations and direct radon gas concentration observations) are needed to
358 improve understanding on the sources of ambient radioactivity in the open ocean and
359 assess whether gamma radiation in the marine environment is influenced not only by ra-
360 dionuclides of terrestrial origin, like radon and its progeny, but also cosmogenic radionu-
361 clides, like Be-7, formed in the upper atmosphere but with the ability to be transported
362 downward and serve as a tracer of the aerosols to which it attaches. This could comple-
363 ment studies of upper troposphere dust sources and transport based on satellite data (Yang
364 et al., 2022), and would improve understanding on planetary environmental radioactiv-
365 ity and the use of radionuclides as tracers of cloud scavenging and precipitation processes,
366 with implications for the use of radionuclides as tracers of transport and residence time
367 of aerosols in the marine boundary layer.

368 5 Open Research

369 Raw measurements from the SAIL campaign are available upon request (S. Bar-
370 bosa et al., 2021). The datasets of processed measurements used in this manuscript are
371 publicly available: gamma radiation data (S. Barbosa, Almeida, et al., 2022a) and vis-
372 ibility data (S. Barbosa, Almeida, et al., 2022b). The analysis was performed using the
373 R software (R Core Team, 2022). Maps were created with the Generic Mapping Tools
374 (GMT) software (Wessel et al., 2019).

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