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1 **Running head:** Mercury contamination of brown skuas

2
3 **Spatial and sex differences in mercury contamination of skuas in the**
4 **Southern Ocean**

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45 **Highlights**

46 • Blood total mercury (THg) concentrations were measured in skuas across a latitudinal
47 gradient.

48 • Hg contamination increased from the Antarctic Peninsula to South Georgia.

49 • Blood THg concentrations showed a weak positive correlation with $\delta^{15}\text{N}$.

50 • Male brown skuas showed higher Hg contamination than females.

51

52 **Abstract:** Antarctic marine ecosystems are often considered to be pristine environments, yet
53 wildlife in the polar regions may still be exposed to high levels of environmental contaminants.

54 Here, we measured total mercury (THg) concentrations in blood samples from adult brown
55 skuas *Stercorarius antarcticus lonnbergi* ($n = 82$) from three breeding colonies south of the

56 Antarctic Polar Front in the Southern Ocean (southwest Atlantic region): (i) Bahía
57 Esperanza/Hope Bay, Antarctic Peninsula; (ii) Signy Island, South Orkney Islands; and, (iii)

58 Bird Island, South Georgia. Blood THg concentrations increased from the Antarctic Peninsula
59 towards the Antarctic Polar Front, such that Hg contamination was lowest at Bahía

60 Esperanza/Hope Bay (mean \pm SD, $0.95 \pm 0.45 \mu\text{g g}^{-1} \text{dw}$), intermediate at Signy Island ($3.42 \pm$
61 $2.29 \mu\text{g g}^{-1} \text{dw}$) and highest at Bird Island ($4.47 \pm 1.10 \mu\text{g g}^{-1} \text{dw}$). Blood THg concentrations

62 also showed a weak positive correlation with $\delta^{15}\text{N}$ values, likely reflecting the biomagnification
63 process. Males had higher Hg burdens than females, which may reflect deposition of Hg into

64 eggs by females or potentially differences in their trophic ecology. These data provide
65 important insights into intraspecific variation in contamination and the geographic transfer of

66 Hg to seabirds in the Southern Ocean.

67

68 **Keywords:** seabird; pollutants; metals; stable isotopes; Antarctica; Southern Hemisphere

69

70

1. Introduction

71 Mercury (Hg) is a pervasive environmental contaminant of major concern for both humans and
72 wildlife. Hg derives from both natural and anthropogenic sources; however, human activities
73 (e.g., coal burning, artisanal and small-scale gold mining) have increased the global Hg pool
74 (Eagles-Smith et al., 2018; UN Environment, 2019). The gaseous, elemental form of Hg (Hg^0)
75 can travel long distances via atmospheric transport to remote locations that are isolated from
76 major emission sources, including the Antarctic (Fitzgerald et al., 1998; Driscoll et al., 2013).
77 Once deposited in the marine environment, inorganic Hg (Hg^{II}) is converted via biotic *in situ*
78 methylation to the more toxic form, methyl-Hg (MeHg, $[\text{CH}_3\text{Hg}]^+$), which, once assimilated,
79 bioaccumulates within marine organisms (i.e., concentrations increase within the body over
80 time) and biomagnifies through marine food webs from lower to higher trophic levels (Bargagli
81 et al., 1998; Blum et al., 2013; Seco et al., 2021). Long-lived, upper trophic level predators,
82 such as many seabirds, are potentially exposed to high levels of Hg through their diets (Cherel
83 et al., 2018).

84 Seabirds are effective bioindicators of Hg contamination in marine ecosystems
85 (Monteiro and Furness, 1995; Monteiro et al., 1996; Carravieri et al., 2016). Hg contamination
86 may also negatively impact aspects of seabird behaviour, physiology and development, and
87 can have short- or long-term fitness consequences (Goutte et al., 2014; Ackerman et al., 2016;
88 Mills et al., 2020). Studies of seabird communities in the Southern Ocean, from the Antarctic
89 continent to the subtropics, have demonstrated extensive interspecific variation in Hg
90 contamination (Anderson et al., 2009a; Carravieri et al., 2014a; Becker et al., 2016; Polito et
91 al., 2016). Hg contamination of seabirds also often differs among conspecifics, and this
92 variation may be driven by factors such as age, sex, breeding status and trophic ecology
93 (Bearhop et al., 2000a; Polito et al., 2016; Mills et al., 2020). Moreover, several studies, mostly
94 undertaken in the Indian Ocean sector of the Southern Ocean, have demonstrated that Hg
95 contamination is lower for seabirds feeding in Antarctic compared to sub-Antarctic and
96 subtropical waters (Carravieri et al., 2014b, 2016, 2017, 2020; Cherel et al., 2018; Mills et al.,
97 2020; Renedo et al., 2020).

98 Stable isotope analysis is a well-established method for studying the trophic ecology of
99 seabirds (Cherel et al., 2000; Phillips et al., 2009, 2011). Stable isotope ratios of carbon
100 ($^{13}\text{C}/^{12}\text{C}$, expressed as $\delta^{13}\text{C}$) and nitrogen ($^{15}\text{N}/^{14}\text{N}$, $\delta^{15}\text{N}$) offer powerful alternatives to

101 conventional diet studies (e.g., analyses of regurgitates or pellets), as stable isotope ratios of
102 seabird tissues (e.g., blood) reflect those of their prey in a predictable way (Peterson and Fry,
103 1987; Hobson and Clark, 1992; Bearhop et al., 2002; Inger and Bearhop, 2008). $\delta^{15}\text{N}$ values
104 increase in a stepwise manner by $\sim 3\text{--}5\text{‰}$ at each trophic level, whereas $\delta^{13}\text{C}$ values increase
105 less with trophic level ($\sim 0.5\text{--}1\text{‰}$) but can be used to infer foraging habitat (Peterson and Fry,
106 1987; Hobson and Clark, 1992; Bearhop et al., 2002; Cherel and Hobson, 2007; Phillips et al.,
107 2011). In the marine environment, this includes the relative reliance on an inshore vs. offshore,
108 benthic vs. pelagic diet, and latitude or water mass where a gradient exists, such as in the
109 Southern Ocean (Cherel and Hobson, 2007; Jaeger et al., 2010; Phillips et al., 2009; Quillfeldt
110 et al., 2005, 2010; St John Glew et al., 2021). $\delta^{15}\text{N}$ values in seabird tissues may also reflect
111 spatial variation in baselines (Elliott et al., 2021). Stable isotope ratios measured in seabird
112 blood samples are appropriate for examining the relationships between Hg contamination and
113 trophic ecology, owing to the broadly similar timeframe for integration into the blood system
114 (Bond, 2010; Albert et al., 2019).

115 This study focuses on Hg contamination of brown skuas *Stercorarius antarcticus*
116 *lonnbergi* from three breeding colonies spanning a large latitudinal gradient in the Southern
117 Ocean (southwest Atlantic region), from the Antarctic Peninsula to South Georgia. Brown
118 skuas are opportunistic predators and scavengers in both marine and terrestrial environments,
119 and use a wide variety of feeding methods including aerial and terrestrial hunting, fishing and
120 kleptoparasitism (Furness, 1987; Phillips et al., 2004; Anderson et al., 2009b; Carneiro et al.,
121 2014). Brown skua diets are variable among populations, for instance, in our study region, the
122 reliance on penguin prey decreases from the Antarctic continent towards South Georgia
123 (Burton, 1968; Reinhardt et al., 2000; Phillips et al., 2004; Anderson et al., 2009b; Graña Grilli
124 and Montalti, 2015; Borghello et al., 2019). Brown skuas also show reversed sexual size
125 dimorphism, with females being significantly larger and heavier than males (Phillips et al.,
126 2002), and Hg burdens are often high in this species owing to their high trophic position (Goutte
127 et al., 2014; Carravieri et al., 2017; Seco et al., 2021). Here, our aim was to analyse how Hg
128 contamination of brown skuas varies among breeding colonies located at different latitudes,
129 between sexes and, using bulk stable isotope ratios as proxies, in relation to trophic ecology.
130 We make the following *a priori* predictions: (i) Hg contamination will increase from the
131 Antarctic Peninsula to lower-latitude breeding colonies; (ii) Hg contamination will be
132 positively correlated with blood $\delta^{15}\text{N}$ values, reflecting the biomagnification process; and (iii)
133 females will show lower levels of Hg contamination than males, potentially reflecting
134 deposition of Hg into eggs (Robinson et al., 2012). Baseline data on Hg contamination is

135 especially important for this species, given the detrimental consequences of high Hg burdens
136 for breeding success elsewhere in the Southern Ocean (Goutte et al., 2014).

137

138 **2. Materials and methods**

139 *2.1. Study area and sample collection*

140 Fieldwork was undertaken at three brown skua breeding colonies spanning a latitudinal gradient
141 south of the Antarctic Polar Front (APF) in the Southern Ocean (southwest Atlantic region):
142 (i) Bahía Esperanza/Hope Bay, Antarctic Peninsula (63°24'S, 57°01'W) in
143 November/December 2019; (ii) Signy Island, South Orkney Islands (60°43'S, 45°36'W) in
144 January 2014; and, (iii) Bird Island, South Georgia (54°00'S, 38°03'W) in December 2014
145 (Fig. 1). At each breeding colony, blood samples were obtained during the mid to late
146 incubation periods from either the tarsal or brachial vein of adult birds using a 25-G needle.
147 Birds return to the breeding colonies a couple of months prior to egg-laying (e.g., most birds
148 return in early to mid-October at Bahía Esperanza/Hope Bay and Bird Island) (Burton, 1968;
149 Carneiro et al., 2016; Ibañez, unpublished data). Blood samples were returned to the laboratory
150 within 2–6 hours. Red blood cells were then isolated via centrifugation and removal of the
151 supernatant, and stored frozen prior to laboratory analyses. Birds were sexed by the analysis of
152 DNA extracted from blood samples at Bird Island, and by weight and tarsus lengths at Bahía
153 Esperanza/Hope Bay and Signy Island (Fridolfsson and Ellegren, 1999; Phillips et al., 2002).
154 The sexes of all birds assigned morphologically at Bahía Esperanza/Hope Bay were later
155 confirmed by DNA analysis.

156

157 *2.2. Total mercury analysis*

158 The vast majority (>90%) of total Hg (THg) in skua blood is MeHg (Renedo et al., 2018), and
159 the estimated half-life of Hg in the blood of great skuas *Stercorarius skua* is 30–60 days
160 (Bearhop et al., 2000b). Blood samples were freeze-dried and homogenised prior to analysis.
161 THg concentrations of red blood cells (hereafter “blood THg”) were measured using an
162 Advanced Mercury Analyser spectrophotometer (Altec AMA 254) at the laboratory Littoral
163 Environnement et Sociétés (LIENSs, France). For each sample, a minimum of two aliquots
164 (range: 0.29–1.86 mg dry weight [dw]) were analysed, and the means and relative standard
165 deviations between measurements were calculated (all samples relative standard deviation
166 <10%). THg concentrations are presented in $\mu\text{g g}^{-1}$ dw. Accuracy was tested using a certified
167 reference material (dogfish liver DOLT-5, NRC, Canada; certified Hg concentration: $0.44 \pm$
168 $0.18 \mu\text{g g}^{-1}$ dw) every 10 samples. The measured values were $0.43 \pm 0.01 \mu\text{g g}^{-1}$ dw ($n = 15$),

169 and hence the recovery was $97.8 \pm 1.7\%$. Blanks were analysed at the beginning of each set of
170 samples and the detection limit of the method was $0.005 \mu\text{g g}^{-1} \text{ dw}$.

171

172 *2.3. Stable isotope analysis*

173 Stable isotope ratios were measured in the same blood samples as above. In great skuas, $\delta^{13}\text{C}$
174 and $\delta^{15}\text{N}$ in blood have half-lives of 15.7 and 14.4 days, respectively (i.e., of similar magnitude
175 to the integration period to Hg, see above) (Bearhop et al., 2002). Stable isotope analyses were
176 conducted at the Natural Environment Isotope Facility (NEIF) Stable Isotope Ecology Lab,
177 SUERC. Stable isotope ratios of carbon and nitrogen were determined by a continuous-flow
178 mass spectrometer (Delta Plus XP; Thermo Scientific, Bremen, Germany) coupled to an
179 elemental analyser (vario PYRO cube; Elementar, Langenselbold, Germany). To correct for
180 instrument drift, three internal laboratory standards were analysed for every 10 samples. Stable
181 isotope ratios are reported as δ -values and expressed as ‰ according to the equation: $\delta X =$
182 $[(R_{\text{sample}}/R_{\text{standard}}) - 1] \times 10^3$, where X is ^{13}C or ^{15}N , R is the corresponding ratio $^{13}\text{C}/^{12}\text{C}$ or
183 $^{15}\text{N}/^{14}\text{N}$, and R_{standard} is the ratio of international references Vienna PeeDee Belemnite for
184 carbon and atmospheric N_2 (AIR) for nitrogen. Measurement precision (standard deviation
185 associated with replicate runs of USGS40) was $<0.2 \text{ ‰}$ for both $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$.

186

187 *2.4. Data analysis*

188 General linear models (GLMs; Gaussian distribution and identity link function) were used to
189 assess variation in log-transformed blood THg concentrations. Predictor variables included sex,
190 breeding colony (three-level factor), blood $\delta^{15}\text{N}$ and blood $\delta^{13}\text{C}$ values. Blood $\delta^{15}\text{N}$ values were
191 included as a predictor variable despite the baseline ^{15}N enrichment with latitude in the
192 Southern Ocean, as this is more pronounced at the Subtropical Front than south of the APF,
193 where our study populations are located (Cherel and Hobson, 2007; Jaeger et al., 2010). Blood
194 $\delta^{13}\text{C}$ values of brown skuas differed significantly among breeding colonies (Kruskal-Wallis
195 test, $H = 67.2$, $p < 0.001$), and so separate models were parameterised with either the breeding
196 colony or $\delta^{13}\text{C}$ as an explanatory variable. Blood $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ values were not highly
197 correlated and were included in the same models (Spearman's correlation, $r_s = 0.34$, $p < 0.01$).
198 Models included relevant two-way interactions among predictor variables and were ranked
199 using the Akaike Information Criteria adjusted for small sample sizes (AIC_c) and models
200 within two AIC_c units of the top model ($\leq 2 \text{ AIC}_c$) were considered equally plausible (Burnham
201 and Anderson, 2002). Akaike weights (ω_i) were used to assess the weight of evidence in favour

202 of a given model among the candidate set (Burnham and Anderson, 2002). All analyses were
203 conducted using R version 3.4.4 (R Core Team, 2019).

204

205

3. Results

206 Blood THg concentrations were measured in 82 individual brown skuas. Mean (\pm SD) blood
207 THg concentrations were $3.13 \pm 2.08 \mu\text{g g}^{-1} \text{ dw}$ (range: $0.49\text{--}8.67 \mu\text{g g}^{-1} \text{ dw}$) (Table 1). The
208 most parsimonious GLM (i.e., $\Delta\text{AIC}_C = 0.00$) explaining variation in log-transformed blood
209 THg concentrations included breeding colony, which reflects an increase in mean blood THg
210 concentrations from Bahía Esperanza/Hope Bay ($0.95 \pm 0.45 \mu\text{g g}^{-1} \text{ dw}$, range: $0.49\text{--}2.33 \mu\text{g g}^{-1} \text{ dw}$,
211 $n = 23$) to Signy Island ($3.42 \pm 2.29 \mu\text{g g}^{-1} \text{ dw}$, range: $1.10\text{--}8.67 \mu\text{g g}^{-1} \text{ dw}$, $n = 28$)
212 and then Bird Island ($4.47 \pm 1.10 \mu\text{g g}^{-1} \text{ dw}$, range: $3.02\text{--}7.46 \mu\text{g g}^{-1} \text{ dw}$, $n = 31$) (Table 1 and
213 Fig. 2). The most parsimonious GLM also contained $\delta^{15}\text{N}$ values, sex and their interaction
214 (Table 2 and Fig. 3A). This reflects a weak positive relationship between blood THg
215 concentrations and blood $\delta^{15}\text{N}$ values, and that although the slopes differed between sexes,
216 mean blood THg concentrations were higher in males than females in all populations (Table 1
217 and Fig. 3A). GLMs replacing the breeding colony variable with blood $\delta^{13}\text{C}$ values produced
218 similar results (Table 1), and indicated a strong positive relationship between blood THg
219 concentrations and blood $\delta^{13}\text{C}$ values (Fig. 3B).

220

221

4. Discussion

222 Antarctic marine ecosystems are often considered to be pristine environments, yet wildlife in
223 the polar regions may still be exposed to high levels of environmental contaminants (Ibañez et
224 al., 2020; Mills et al., 2020). This study provides a detailed evaluation of the underlying drivers
225 of blood THg contamination of brown skuas from three breeding colonies located south of the
226 APF in the Antarctic Zone of the Southern Ocean. Blood samples provide a non-lethal means
227 of obtaining information about Hg contamination of seabirds, and measuring THg in skua
228 blood samples provides information about contamination of prey and hence exposure to MeHg
229 in food webs (Albert et al., 2019).

230

231

4.1. Latitudinal differences: from the Antarctic Peninsula to South Georgia

232 Our analysis demonstrates clear differences in Hg contamination of brown skuas among the
233 three breeding colonies located at different latitudes south of the APF. Blood THg
234 concentrations were lowest at the Antarctic Peninsula, intermediate at the South Orkney Islands
235 and highest at South Georgia. Indeed, mean blood THg concentrations were 4.7x higher at Bird

236 Island compared to Bahía Esperanza/Hope Bay. This pattern is corroborated by the strong
237 positive correlation between THg and $\delta^{13}\text{C}$ values (Fig. 3B), as $\delta^{13}\text{C}$ increases from the
238 Antarctic continent towards the subtropics in the Southern Ocean (Cherel and Hobson, 2007;
239 Jaeger et al., 2010; Quillfeldt et al., 2005, 2010; Phillips et al. 2009; St John Glew et al., 2021).
240 Hence, our study suggests geographic differences in the transfer of Hg to skuas in the southwest
241 Atlantic sector of the Southern Ocean, and our first *a priori* prediction is supported. This result
242 contrasts with other studies concluding that breeding latitude has a limited effect on Hg
243 contamination of seabirds in waters south of the APF (Cherel et al., 2018; Carravieri et al.,
244 2020). Nonetheless, a pattern of increasing Hg contamination of seabirds from Antarctic waters
245 towards the sub-Antarctic and subtropics is commonly observed in the Southern Hemisphere
246 (Carravieri et al., 2014b, 2016, 2017, 2020; Cherel et al., 2018; Mills et al., 2020; Renedo et
247 al., 2020). For instance, blood THg concentrations of skua chicks in the southern Indian Ocean
248 increase from breeding sites in the Antarctic to those in the sub-Antarctic and subtropics, with
249 skuas at Amsterdam Island showing the highest recorded concentrations for seabird chicks
250 globally ($3.98 \pm 0.77 \mu\text{g g}^{-1} \text{ dw}$) (Carravieri et al., 2017; Table 3). The majority of MeHg
251 accumulated by seabirds has a mesopelagic origin, and it has been suggested that more efficient
252 methylation of Hg at depth at lower compared to higher latitudes (i.e., subtropical compared to
253 Antarctic waters), combined with higher vertical mixing, could deliver newly formed MeHg to
254 the surface and therefore determine the regional differences in the bioavailability to seabirds
255 (Renedo et al., 2020). To our knowledge, birds from Bahía Esperanza/Hope Bay show the
256 lowest mean blood THg concentrations among adults of all large skua species for which data
257 are available (Table 3); whereas blood THg concentrations at Bird Island were only lower than
258 adults from Kerguelen, and great skuas from St Kilda (Table 3).

259 Differences in Hg contamination among the three breeding colonies may also be
260 exacerbated by dietary differences. Blood $\delta^{13}\text{C}$ values suggest that not all brown skuas from
261 our study populations feed exclusively on Antarctic prey during the incubation and pre-laying
262 periods (i.e., the timeframe reflected by blood $\delta^{13}\text{C}$ values), and feed to some degree on prey
263 from warmer waters north of the APF (Fig. 3B). At Bird Island, brown skuas mainly undertake
264 coastal foraging trips during incubation and feed on the placentae from Antarctic fur seals
265 *Arctocephalus gazella*, supplemented by carrion, particularly male seals killed during territorial
266 conflicts (Phillips et al., 2004; Anderson et al., 2009b; Carneiro et al., 2014). Many of the
267 female fur seals would have spent the winter foraging north of the APF, and potentially over
268 the Patagonian Shelf (Staniland et al., 2012). Conversely, pellet analyses indicate that penguins
269 are the dominant prey consumed at the higher latitude colonies (Signy Island and Bahía

270 Esperanza/Hope Bay) (Burton, 1968; Graña Grilli and Montalti, 2015; Borghello et al., 2019;
271 Ibañez et al., unpublished data), which forage in Antarctic waters (Lynnes et al., 2002). Indeed,
272 the brown skuas at Bahía Esperanza/Hope Bay breed in close proximity to a large Adélie
273 penguin *Pygoscelis adeliae* colony (approximately 100,000 breeding pairs), and two smaller
274 gentoo penguin *P. papua* colonies (Santos et al., 2018; Borghello et al., 2019). Another
275 contributing factor is that birds may undertake a pre-laying exodus after returning to the colony.
276 A considerable proportion of brown skuas from Bird Island perform a pre-laying exodus north
277 to sub-Antarctic and mixed sub-Antarctic-subtropical waters where the bioavailability of Hg is
278 relatively high (Phillips et al., 2007; Carneiro et al., 2016a). Unfortunately, the pre-laying
279 exodus is not well described at other colonies, including at the higher latitude breeding colonies
280 studied here. Prey ingested around the colony during breeding will therefore represent the
281 primary influence on blood THg in skuas; however, there may be a weak carry-over effect of
282 Hg exposure during winter, and hence differing migration strategies may also have contributed
283 to the patterns observed here.

284

285 4.2. Relationships with blood $\delta^{15}\text{N}$ values

286 The most parsimonious GLMs explaining blood THg concentrations included blood $\delta^{15}\text{N}$
287 values as a predictor variable, which provide a proxy for trophic position, and supports our
288 second *a priori* prediction. Although the slopes differed between sexes, the weak positive
289 correlation between blood THg and $\delta^{15}\text{N}$ values is likely to reflect, at least in part, the
290 biomagnification of MeHg within marine food webs, such that individuals foraging at higher
291 trophic levels were exposed to greater levels of Hg. However, the positive relationships
292 between blood THg and $\delta^{13}\text{C}$ values were stronger than with $\delta^{15}\text{N}$ values (Fig. 3A and Fig. 3B).
293 Biomagnification of Hg has been demonstrated in the Scotia Sea ecosystem, from particulate
294 organic matter through to higher trophic levels (Anderson et al., 2009a; Seco et al., 2021).
295 Indeed, mean blood THg concentrations of several large seabird species in our study region are
296 higher than those of brown skuas in this study; however, it is notable that those of southern
297 giant petrels *Macronectes giganteus* ($2.74 \pm 1.05 \mu\text{g g}^{-1}$ dw, range: 1.52–4.74 $\mu\text{g g}^{-1}$ dw), and
298 northern giant petrels *M. halli* ($3.93 \pm 1.37 \mu\text{g g}^{-1}$ dw, range: 2.18–6.38 $\mu\text{g g}^{-1}$ dw) from Bird
299 Island in 2001/02 were lower than in brown skuas from Bird Island in 2014/15 (Anderson et
300 al., 2009a; this study). The relationship between $\delta^{15}\text{N}$ and Hg contamination is often apparent
301 when comparing values among different species within seabird communities (e.g., Blévin et
302 al., 2013; Anderson et al., 2009a), but is observed less frequently within a single species, and
303 was apparent across our study populations despite relatively little variation in $\delta^{15}\text{N}$ values

304 (range: 10.2 to 12.8 ‰). Alternatively, because there is a baseline ^{15}N enrichment from the
305 Antarctic towards the subtropics in the Southern Ocean (Cherel and Hobson, 2007; Jaeger et
306 al., 2010), the relationship between Hg and $\delta^{15}\text{N}$ may simply reflect the increasing
307 contamination with latitude (i.e., trophic variation may be obscured by spatial variation).
308 However, changes in baseline $\delta^{15}\text{N}$ are less pronounced south of the APF (Cherel and Hobson,
309 2007; Jaeger et al., 2010). Moreover, differences in mean blood $\delta^{15}\text{N}$ values among populations
310 were small (means at Bird Island are ~ 0.2 ‰ higher than at Bahía Esperanza/Hope Bay; Table
311 1), and mean $\delta^{15}\text{N}$ values were slightly lower at Signy Island than at Bahía Esperanza/Hope
312 Bay (Table 1).

313

314

4.3. Sex differences in Hg contamination

315 Sex was also included as a predictor variable in the most parsimonious GLM, and blood THg
316 was lower in females than males at each breeding colony (Table 1). Hence, our third *a priori*
317 prediction is supported. As the blood samples were taken in mid to late incubation, two non-
318 mutually exclusive hypotheses may explain the sex differences. First, egg production offers a
319 route by which females are able to eliminate Hg (Bond and Diamond, 2008; Robinson et al.,
320 2012; Ackerman et al., 2020), and Hg in eggs is thought to reflect dietary intake by females in
321 the period before laying (Lewis et al., 1993; Monteiro and Furness, 1995). Second, our data
322 show slight sex differences in blood $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ values for all three populations (Table 1);
323 hence, there may also be trophic differences between sexes. At Bird Island at least, females are
324 more likely than males to undertake a pre-laying exodus (Phillips et al., 2007; Carneiro et al.,
325 2016a). However, during late incubation and early chick-rearing at Bird Island, the sexes show
326 similar territorial attendance, foraging time and habitat use, and a previous stable isotope study
327 at South Georgia found no evidence for sex-specific foraging preferences during the breeding
328 period (Anderson et al., 2009b; Carneiro et al., 2014). This is despite pronounced reverse sexual
329 size dimorphism in this species (Catry et al., 1999; Phillips et al., 2002).

330

331

4.4. Could Hg contamination have fitness implications?

332 Hg is a neurotoxin and endocrine disruptor (Tan et al., 2009), and contamination may impact
333 various aspects of development, physiology and breeding behaviour in seabirds (Burger and
334 Gochfeld, 1997; Evers et al., 2008; Tartu et al., 2013; Goutte et al., 2015). Hg contamination
335 has also been associated with reduced breeding success (Goutte et al., 2014; Ackerman et al.,
336 2016; Mills et al., 2020). The mean blood THg (dw) concentrations reported here are equivalent
337 to $0.94 \mu\text{g g}^{-1}$ wet weight (ww), $0.72 \mu\text{g g}^{-1}$ ww and $0.20 \mu\text{g g}^{-1}$ ww at Bird Island, Signy Island

338 and Bahía Esperanza/Hope Bay, respectively (assuming a 79% moisture content) (Eagles-
339 Smith et al., 2008; Ackerman et al., 2016). These values suggest a low risk of MeHg toxicity
340 (Ackerman et al., 2016). Indeed, mean blood THg concentrations in our study were far lower
341 than those of brown skuas at Kerguelen; however, blood THg concentrations at Bird Island and
342 Signy Island exceeded those of south polar skuas *Stercorarius maccormicki* at Adélie Land
343 (Goutte et al., 2014; Table 3). Goutte et al. (2014) found short-term associations between Hg
344 contamination and breeding success in both skua species, yet long-term reproductive impacts
345 were more severe in south polar skuas than brown skuas, despite their lower blood THg
346 concentrations. Selenium (Se) offers a well-known protective effect against Hg toxicity when
347 in molar excess (i.e., when Se:Hg > 1), and blood Se:Hg molar ratios of south polar skuas were
348 three times lower than the brown skuas (Carravieri et al., 2017; Manceau et al., 2021). Hence,
349 although it currently appears unlikely that there are fitness consequences of Hg contamination
350 for brown skuas in our study region (though we did not test this directly), the role of Se requires
351 evaluation. Populations of brown skuas at Signy Island and Bird Island increased rapidly from
352 the late-1950s to early-1980s (by 3.3% and 3.6% pa, respectively), but then rates slowed until
353 the 2010s (0.4% pa) and 2000s (0.9% pa), respectively (Phillips et al., 2004; Carneiro et al.,
354 2016b). Rather than a consequence of Hg contamination, the slower population growth rates
355 seem more likely to be related to changes in prey availability and increased density-dependent
356 competition (Phillips et al., 2004; Carneiro et al., 2016b).

357

358

5. Conclusion

359 Our study provides important baseline data on Hg contamination of brown skuas in the
360 Southern Ocean, and analyses the underlying drivers of variation in contamination. Given the
361 relatively high levels of Hg contamination of brown skuas at Bird Island, which in the early
362 2000s was likely the most densely-populated colony in the world (132 nesting pairs per km² of
363 suitable habitat) (Phillips et al., 2004), further monitoring is required, particularly as there is
364 some evidence for increasing Hg contamination of other seabirds at South Georgia (Mills et
365 al., 2020). Moreover, MeHg concentrations peak in low-oxygen subsurface waters, and in a
366 warming world, oxygen minimum zones are expected to increase, hence potentially enhancing
367 methylation of Hg and its bioavailability to marine predators (Blum et al., 2013). Further work
368 should also examine the potential protective effects of Se on Hg toxicity in brown skuas, and
369 the risks posed by other contaminants to these skua populations, especially given the incidence
370 of plastic ingestion by this species at the Antarctic Peninsula and the potential role of plastics
371 in contaminant transmission (Lavers and Bond, 2016; Ibañez et al., 2020).

372

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Declaration of competing interest

374 The authors declare that they have no known competing financial interests or personal
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CRedit authorship statement

394 WFM: Conceptualization, Methodology, Investigation, Formal Analysis, Visualization,
395 Writing - Original Draft. AEI: Conceptualization, Methodology, Investigation, Resources,
396 Writing - Review & Editing. PB: Investigation, Writing - Review & Editing. APBC: Resources,
397 Investigation, Writing - Review & Editing. SB: Supervision, Writing - Review & Editing. YC:
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399 & Editing. RARM: Investigation, Writing - Review & Editing. DM: Supervision, Writing -
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Table 1. Total mercury (THg) concentrations ($\mu\text{g g}^{-1}$ dw) in red blood cells sampled from adult brown skuas *Stercorarius antarcticus lonnbergi* at: (i) Bahía Esperanza/Hope Bay, Antarctic Peninsula ($63^{\circ}24'S$, $57^{\circ}01'W$); (ii) Signy Island, South Orkney Islands ($60^{\circ}43'S$, $45^{\circ}36'W$); and, (iii) Bird Island, South Georgia ($54^{\circ}00'S$, $38^{\circ}03'W$). Values are means \pm SDs.

Breeding population	Year	<i>n</i>	Sex	THg ($\mu\text{g g}^{-1}$ dw)	$\delta^{15}\text{N}$ (‰)	$\delta^{13}\text{C}$ (‰)
Bahía Esperanza/Hope Bay, Antarctic Peninsula	2019	12	Male	1.02 ± 0.50	11.61 ± 0.49	-23.26 ± 0.26
		11	Female	0.87 ± 0.40	11.25 ± 0.53	-23.06 ± 0.41
		23	Both	0.95 ± 0.45	11.44 ± 0.53	-23.17 ± 0.35
Signy Island, South Orkney Islands	2014	15	Male	3.92 ± 2.30	11.44 ± 0.48	-22.29 ± 0.52
		13	Female	2.85 ± 2.23	11.24 ± 0.65	-22.10 ± 0.39
		28	Both	3.42 ± 2.29	11.35 ± 0.56	-22.20 ± 0.47
Bird Island, South Georgia	2015	14	Male	4.86 ± 1.12	11.81 ± 0.51	-20.17 ± 0.22
		17	Female	4.15 ± 1.00	11.50 ± 0.61	-20.31 ± 0.29
		31	Both	4.47 ± 1.10	11.64 ± 0.58	-20.24 ± 0.26

Table 2. Model selection for factors explaining variation in log-transformed total mercury (THg) concentrations ($\mu\text{g g}^{-1}$ dw) of red blood cells from adult brown skuas *Stercorarius antarcticus lonnbergi*. The top 4 models are shown, and all are GLMs (Gaussian distribution and identity link function). k is the number of parameters; ΔAIC_c is the change in the Akaike information criterion corrected for small sample sizes (AIC_c) from the best-supported model; ω_i is the Akaike weight; Exp. Dev. is the percentage explained deviance of the top model.

Models	k	ΔAIC_c	ω_i^a	Exp. Dev. (%)
<hr/> Maximal model: $\text{THg} \sim \text{Sex} + \text{Site} + \delta^{15}\text{N} + \text{Sex}:\delta^{15}\text{N} + \text{Site}:\text{Sex} + \text{Site}:\delta^{15}\text{N}$ <hr/>				
$\text{THg} \sim \text{Sex} + \text{Site} + \delta^{15}\text{N} + \text{Sex}:\delta^{15}\text{N}$	7	0.00	0.57	79.1
$\text{THg} \sim \text{Sex} + \text{Site} + \delta^{15}\text{N} + \text{Sex}:\delta^{15}\text{N} + \text{Site}:\text{Sex}$	9	2.32	0.18	
$\text{THg} \sim \text{Sex} + \text{Site} + \delta^{15}\text{N} + \text{Sex}:\delta^{15}\text{N} + \text{Site}:\delta^{15}\text{N}$	9	2.83	0.14	
$\text{THg} \sim \text{Sex} + \text{Site} + \delta^{15}\text{N} + \text{Sex}:\delta^{15}\text{N} + \text{Site}:\text{Sex} + \text{Site}:\delta^{15}\text{N}$	13	5.44	0.04	
<hr/> Maximal model ^b : $\text{THg} \sim \text{Sex} + \delta^{13}\text{C} + \delta^{15}\text{N} + \text{Sex}:\delta^{13}\text{C} + \text{Sex}:\delta^{15}\text{N}$ <hr/>				
$\text{THg} \sim \text{Sex} + \delta^{13}\text{C} + \delta^{15}\text{N} + \text{Sex}:\delta^{15}\text{N}$	6	0.00	0.63	66.1
$\text{THg} \sim \text{Sex} + \delta^{13}\text{C} + \delta^{15}\text{N} + \text{Sex}:\delta^{13}\text{C} + \text{Sex}:\delta^{15}\text{N}$	7	2.22	0.21	
$\text{THg} \sim \text{Sex} + \delta^{13}\text{C}$	4	4.70	0.06	
$\text{THg} \sim \text{Sex} + \delta^{13}\text{C} + \text{Sex}:\delta^{13}\text{C}$	5	4.85	0.06	

^aWeights across all models sum to 1.00 (not all shown).

^bWe did not consider an interaction between blood $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values.

Table 3. Review of published blood total mercury (THg) concentrations ($\mu\text{g g}^{-1}$ dw) in adults and chicks of large skua species in the Northern and Southern Hemispheres. Values are means \pm SDs.

Skua species	Sampling year(s)	Age	<i>n</i>	Breeding site	THg ($\mu\text{g g}^{-1}$ dw)	References
Northern hemisphere						
Great skua <i>Stercorarius skua</i>	1996	Chick	27	Foula, Shetland	0.27 ± 0.16	Bearhop et al. (2000c)
	1997	Chick	23	St Kilda, Western Isles	1.12 ± 0.41	Bearhop et al. (2000c)
	1996	Adult	57	Foula, Shetland	3.49 ± 1.83	Bearhop et al. (2000a)
	1996	Adult	28	Foula, Shetland	3.67 ± 2.03	Bearhop et al. (2000c)
	1996–1997	Adult	33	St Kilda, Western Isles	6.71 ± 3.08	Bearhop et al. (2000a)
	1997	Adult	23	St Kilda, Western Isles	7.37 ± 3.10	Bearhop et al. (2000c)
Southern Hemisphere						
Brown skua <i>Stercorarius antarcticus lonnbergi</i>	2011	Chick	10	Kerguelen archipelago	2.31 ± 0.33	Carravieri et al. (2017)
	2011	Chick	10	Kerguelen archipelago	2.32 ± 0.34	Renedo et al. (2020)
	2011	Chick	10	Crozet archipelago	1.66 ± 1.20	Carravieri et al. (2017)
	2012	Chick	11	Crozet archipelago	1.81 ± 1.19	Renedo et al. (2020)
	2011	Chick	10	Amsterdam Island	3.98 ± 0.77	Carravieri et al. (2017)
	2011	Chick	10	Amsterdam Island	4.00 ± 0.89	Renedo et al. (2020)
	2019	Adult	23	Hope Bay, Antarctic Peninsula	0.95 ± 0.45	This study
	2014	Adult	28	Signy Island, South Orkney Islands	3.42 ± 2.29	This study
	2014	Adult	31	Bird Island, South Georgia	4.47 ± 1.10	This study
	2001	Adult	68	Kerguelen archipelago	8.22 ± 1.98	Goutte et al. (2014)
South polar skua <i>Stercorarius maccormicki</i>	2011	Chick	11	Adélie Land, Antarctica	0.51 ± 0.1	Carravieri et al. (2017)
	2011–2012	Chick	9	Adélie Land, Antarctica	0.53 ± 0.08	Renedo et al. (2020)
	2003–2005	Adult	76	Adélie Land, Antarctica	2.15 ± 1.48	Goutte et al. (2014)
	2019	Adult	6	Hope Bay, Antarctic Peninsula	1.41 ± 1.01	Mills, unpublished data

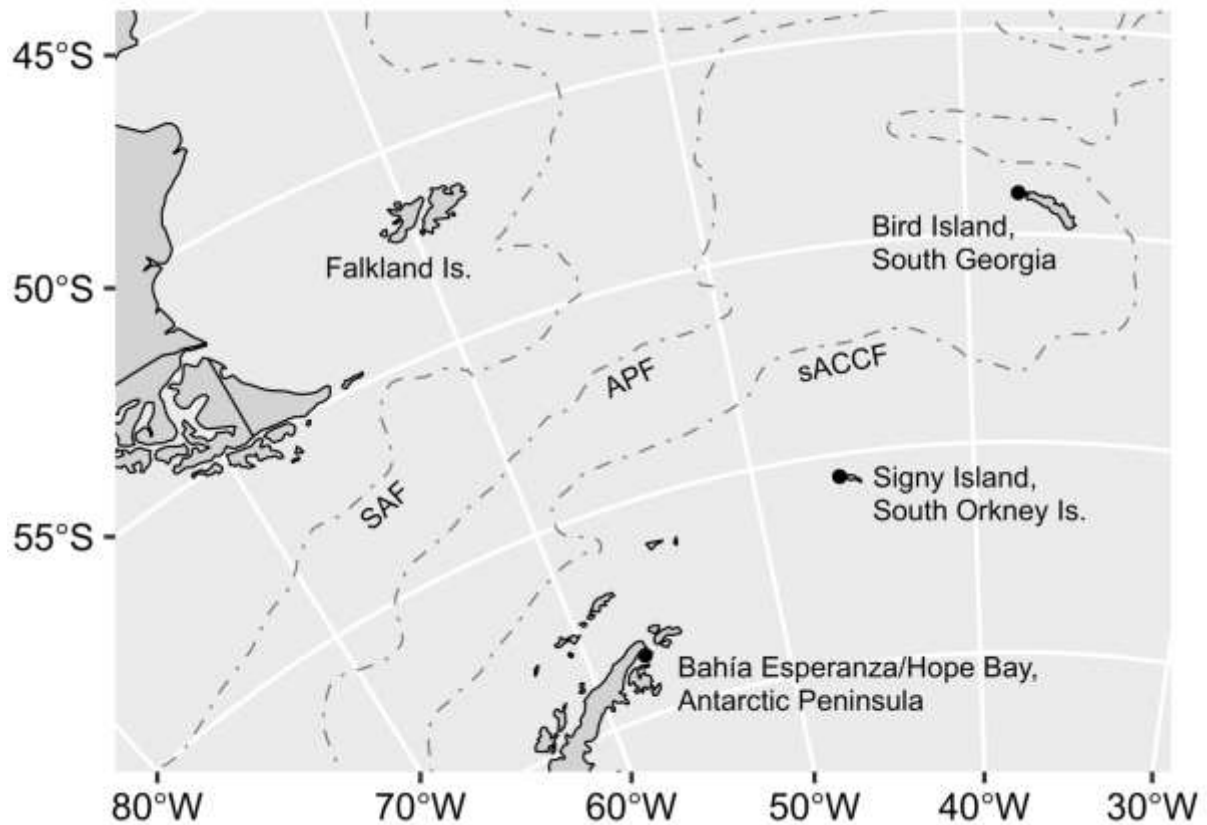


Figure 1. Location of the three study sites in relation to the Sub-Antarctic Front (SAF), the Antarctic Polar Front (APF) and the southern Antarctic Circumpolar Current Front (sACCF). The study sites are: (i) Bahía Esperanza/Hope Bay, Antarctic Peninsula ($63^{\circ}24'S$, $57^{\circ}01'W$); (ii) Signy Island, South Orkney Islands ($60^{\circ}43'S$, $45^{\circ}36'W$); and (iii) Bird Island, South Georgia ($54^{\circ}00'S$, $38^{\circ}03'W$).

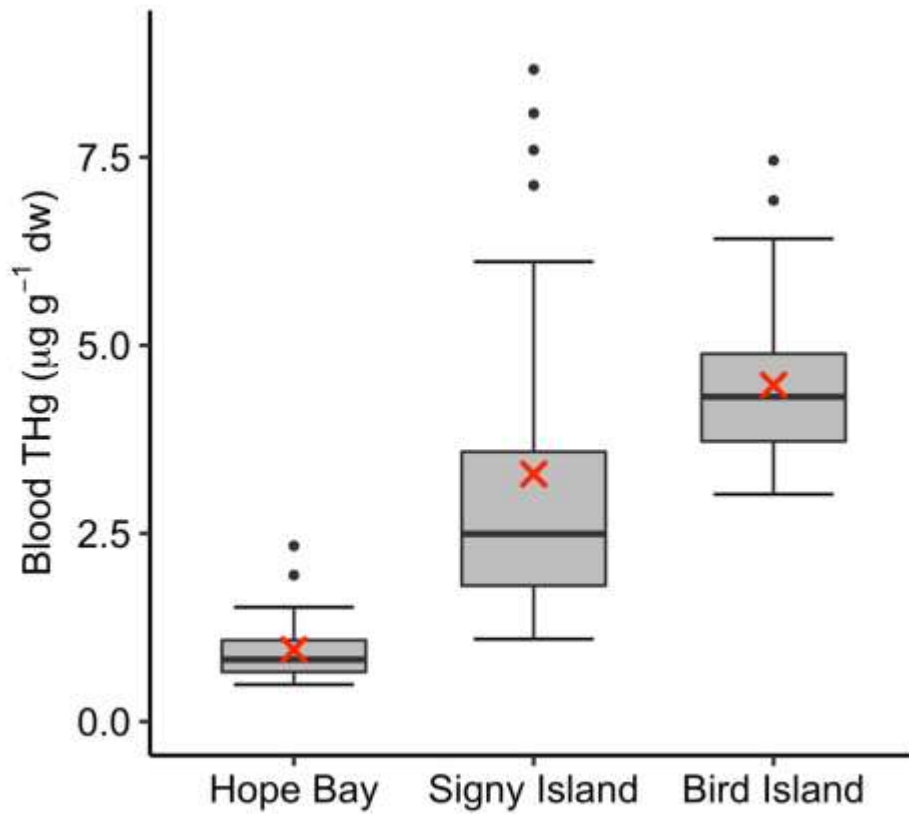


Figure 2. Box plots showing significant differences in the total mercury (THg) concentrations ($\mu\text{g g}^{-1} \text{ dw}$) of red blood cells sampled from adult brown skuas *Stercorarius antarcticus lonnbergi* at: Bahía Esperanza/Hope Bay, Antarctic Peninsula ($63^{\circ}24'S$, $57^{\circ}01'W$); Signy Island, South Orkney Islands ($60^{\circ}43'S$, $45^{\circ}36'W$); and, Bird Island, South Georgia ($54^{\circ}00'S$, $38^{\circ}03'W$). Red crosses are mean values.

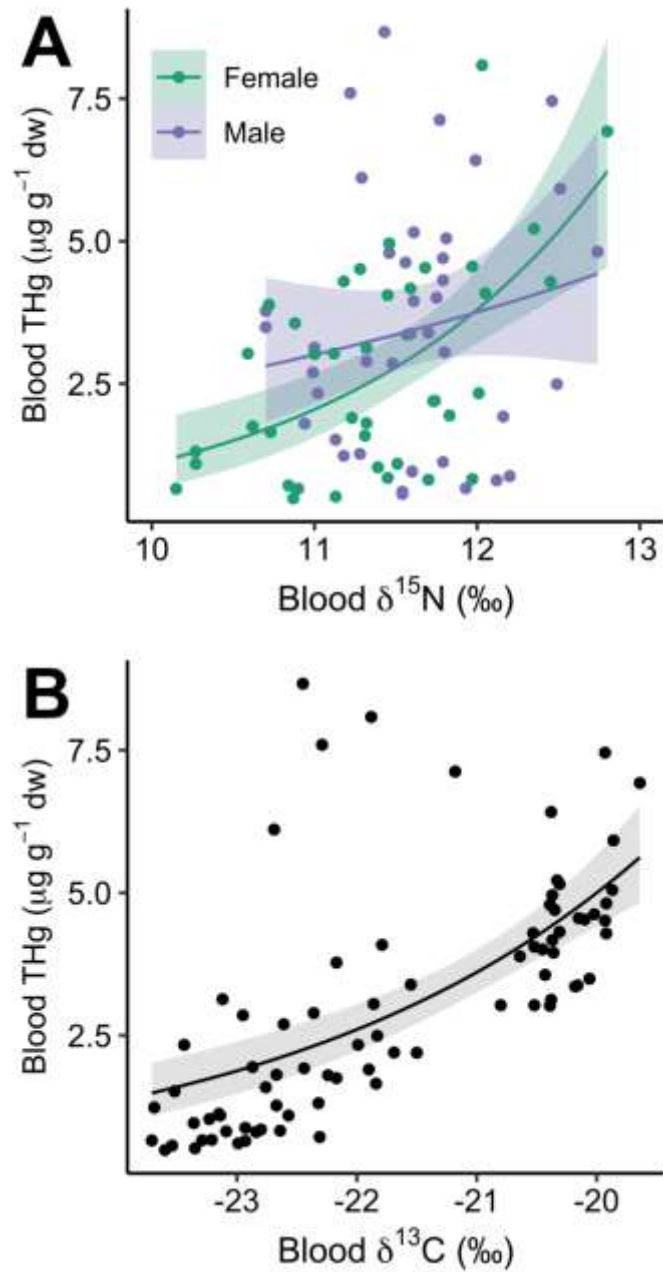


Figure 3. Relationships between total mercury (THg) concentrations ($\mu\text{g g}^{-1} \text{dw}$) and bulk stable isotope ratios of (A) nitrogen ($\delta^{15}\text{N}$), and (B) carbon ($\delta^{13}\text{C}$) in red blood cells sampled from adult brown skuas *Stercorarius antarcticus lonnbergi* from three breeding colonies: Bahía Esperanza/Hope Bay, Antarctic Peninsula ($63^{\circ}24'\text{S}$, $57^{\circ}01'\text{W}$); Signy Island, South Orkney Islands ($60^{\circ}43'\text{S}$, $45^{\circ}36'\text{W}$); and, Bird Island, South Georgia ($54^{\circ}00'\text{S}$, $38^{\circ}03'\text{W}$).