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# **Spatial and sex differences in mercury contamination of skuas in the Southern Ocean**

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## Highlights

- Blood total mercury (THg) concentrations were measured in skuas across a latitudinal gradient.
- Hg contamination increased from the Antarctic Peninsula to South Georgia.
- Blood THg concentrations showed a weak positive correlation with  $\delta^{15}\text{N}$ .
- Male brown skuas showed higher Hg contamination than females.

**Abstract:** Antarctic marine ecosystems are often considered to be pristine environments, yet wildlife in the polar regions may still be exposed to high levels of environmental contaminants. Here, we measured total mercury (THg) concentrations in blood samples from adult brown skuas *Stercorarius antarcticus lonnbergi* ( $n = 82$ ) from three breeding colonies south of the Antarctic Polar Front in the Southern Ocean (southwest Atlantic region): (i) Bahía Esperanza/Hope Bay, Antarctic Peninsula; (ii) Signy Island, South Orkney Islands; and, (iii) Bird Island, South Georgia. Blood THg concentrations increased from the Antarctic Peninsula towards the Antarctic Polar Front, such that Hg contamination was lowest at Bahía Esperanza/Hope Bay (mean  $\pm$  SD,  $0.95 \pm 0.45 \mu\text{g g}^{-1}$  dw), intermediate at Signy Island ( $3.42 \pm 2.29 \mu\text{g g}^{-1}$  dw) and highest at Bird Island ( $4.47 \pm 1.10 \mu\text{g g}^{-1}$  dw). Blood THg concentrations also showed a weak positive correlation with  $\delta^{15}\text{N}$  values, likely reflecting the biomagnification process. Males had higher Hg burdens than females, which may reflect deposition of Hg into eggs by females or potentially differences in their trophic ecology. These data provide important insights into intraspecific variation in contamination and the geographic transfer of Hg to seabirds in the Southern Ocean.

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

## 1. Introduction

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

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

Stable isotope analysis is a well-established method for studying the trophic ecology of seabirds (Cherel et al., 2000; Phillips et al., 2009, 2011). Stable isotope ratios of carbon ( $^{13}\text{C}/^{12}\text{C}$ , expressed as  $\delta^{13}\text{C}$ ) and nitrogen ( $^{15}\text{N}/^{14}\text{N}$ ,  $\delta^{15}\text{N}$ ) offer powerful alternatives to conventional diet studies (e.g., analyses of regurgitates or pellets), as stable isotope ratios of seabird tissues (e.g., blood) reflect those of their prey in a predictable way (Peterson and Fry,

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

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

## 2. Materials and methods

### 2.1. Study area and sample collection

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

### 2.2. Total mercury analysis

The vast majority (>90%) of total Hg (THg) in skua blood is MeHg (Renedo et al., 2018), and the estimated half-life of Hg in the blood of great skuas *Stercorarius skua* is 30–60 days (Bearhop et al., 2000b). Blood samples were freeze-dried and homogenised prior to analysis. THg concentrations of red blood cells (hereafter “blood THg”) were measured using an Advanced Mercury Analyser spectrophotometer (Altec AMA 254) at the laboratory Littoral Environnement et Sociétés (LIENSs, France). For each sample, a minimum of two aliquots (range: 0.29–1.86 mg dry weight [dw]) were analysed, and the means and relative standard deviations between measurements were calculated (all samples relative standard deviation <10%). THg concentrations are presented in  $\mu\text{g g}^{-1}$  dw. Accuracy was tested using a certified reference material (dogfish liver DOLT-5, NRC, Canada; certified Hg concentration:  $0.44 \pm 0.18 \mu\text{g g}^{-1}$  dw) every 10 samples. The measured values were  $0.43 \pm 0.01 \mu\text{g g}^{-1}$  dw ( $n = 15$ ), and hence the recovery was  $97.8 \pm 1.7\%$ . Blanks were analysed at the beginning of each set of samples and the detection limit of the method was  $0.005 \mu\text{g g}^{-1}$  dw.

### 2.3. Stable isotope analysis

Stable isotope ratios were measured in the same blood samples as above. In great skuas,  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  in blood have half-lives of 15.7 and 14.4 days, respectively (i.e., of similar magnitude to the integration period to Hg, see above) (Bearhop et al., 2002). Stable isotope analyses were conducted at the Natural Environment Isotope Facility (NEIF) Stable Isotope Ecology Lab, SUERC. Stable isotope ratios of carbon and nitrogen were determined by a continuous-flow mass spectrometer (Delta Plus XP; Thermo Scientific, Bremen, Germany) coupled to an elemental analyser (vario PYRO cube; Elementar, Langenselbold, Germany). To correct for instrument drift, three internal laboratory standards were analysed for every 10 samples. Stable isotope ratios are reported as  $\delta$ -values and expressed as ‰ according to the equation:  $\delta X = [(R_{\text{sample}}/R_{\text{standard}}) - 1] \times 10^3$ , where  $X$  is  $^{13}\text{C}$  or  $^{15}\text{N}$ ,  $R$  is the corresponding ratio  $^{13}\text{C}/^{12}\text{C}$  or  $^{15}\text{N}/^{14}\text{N}$ , and  $R_{\text{standard}}$  is the ratio of international references Vienna PeeDee Belemnite for carbon and atmospheric  $\text{N}_2$  (AIR) for nitrogen. Measurement precision (standard deviation associated with replicate runs of USGS40) was  $<0.2$  ‰ for both  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$ .

### 2.4. Data analysis

General linear models (GLMs; Gaussian distribution and identity link function) were used to assess variation in log-transformed blood THg concentrations. Predictor variables included sex, breeding colony (three-level factor), blood  $\delta^{15}\text{N}$  and blood  $\delta^{13}\text{C}$  values. Blood  $\delta^{15}\text{N}$  values were included as a predictor variable despite the baseline  $^{15}\text{N}$  enrichment with latitude in the Southern Ocean, as this is more pronounced at the Subtropical Front than south of the APF, where our study populations are located (Cherel and Hobson, 2007; Jaeger et al., 2010). Blood  $\delta^{13}\text{C}$  values of brown skuas differed significantly among breeding colonies (Kruskal-Wallis test,  $H = 67.2$ ,  $p < 0.001$ ), and so separate models were parameterised with either the breeding colony or  $\delta^{13}\text{C}$  as an explanatory variable. Blood  $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$  values were not highly correlated and were included in the same models (Spearman's correlation,  $r_s = 0.34$ ,  $p < 0.01$ ). Models included relevant two-way interactions among predictor variables and were ranked using the Akaike Information Criteria adjusted for small sample sizes ( $\text{AIC}_c$ ) and models within two  $\text{AIC}_c$  units of the top model ( $\leq 2 \text{AIC}_c$ ) were considered equally plausible (Burnham and Anderson, 2002). Akaike weights ( $\omega_i$ ) were used to assess the weight of evidence in favour of a given model among the candidate set (Burnham and Anderson, 2002). All analyses were conducted using R version 3.4.4 (R Core Team, 2019).

### 3. Results

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

### 4. Discussion

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

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

Our analysis demonstrates clear differences in Hg contamination of brown skuas among the three breeding colonies located at different latitudes south of the APF. Blood THg concentrations were lowest at the Antarctic Peninsula, intermediate at the South Orkney Islands and highest at South Georgia. Indeed, mean blood THg concentrations were 4.7x higher at Bird Island compared to Bahía Esperanza/Hope Bay. This pattern is corroborated by the strong positive correlation between THg and  $\delta^{13}\text{C}$  values (Fig. 3B), as  $\delta^{13}\text{C}$  increases from the Antarctic continent towards the subtropics in the Southern Ocean (Cherel and Hobson, 2007;



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

Differences in Hg contamination among the three breeding colonies may also be exacerbated by dietary differences. Blood  $\delta^{13}\text{C}$  values suggest that not all brown skuas from our study populations feed exclusively on Antarctic prey during the incubation and pre-laying periods (i.e., the timeframe reflected by blood  $\delta^{13}\text{C}$  values), and feed to some degree on prey from warmer waters north of the APF (Fig. 3B). At Bird Island, brown skuas mainly undertake coastal foraging trips during incubation and feed on the placentae from Antarctic fur seals *Arctocephalus gazella*, supplemented by carrion, particularly male seals killed during territorial conflicts (Phillips et al., 2004; Anderson et al., 2009b; Carneiro et al., 2014). Many of the female fur seals would have spent the winter foraging north of the APF, and potentially over the Patagonian Shelf (Staniland et al., 2012). Conversely, pellet analyses indicate that penguins are the dominant prey consumed at the higher latitude colonies (Signy Island and Bahía Esperanza/Hope Bay) (Burton, 1968; Graña Grilli and Montalti, 2015; Borghello et al., 2019; Ibañez et al., unpublished data), which forage in Antarctic waters (Lynnes et al., 2002). Indeed, the brown skuas at Bahía Esperanza/Hope Bay breed in close proximity to a large Adélie

penguin *Pygoscelis adeliae* colony (approximately 100,000 breeding pairs), and two smaller gentoo penguin *P. papua* colonies (Santos et al., 2018; Borghello et al., 2019). Another contributing factor is that birds may undertake a pre-laying exodus after returning to the colony. A considerable proportion of brown skuas from Bird Island perform a pre-laying exodus north to sub-Antarctic and mixed sub-Antarctic-subtropical waters where the bioavailability of Hg is relatively high (Phillips et al., 2007; Carneiro et al., 2016a). Unfortunately, the pre-laying exodus is not well described at other colonies, including at the higher latitude breeding colonies studied here. Prey ingested around the colony during breeding will therefore represent the primary influence on blood THg in skuas; however, there may be a weak carry-over effect of Hg exposure during winter, and hence differing migration strategies may also have contributed to the patterns observed here.

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

The most parsimonious GLMs explaining blood THg concentrations included blood  $\delta^{15}\text{N}$  values as a predictor variable, which provide a proxy for trophic position, and supports our second *a priori* prediction. Although the slopes differed between sexes, the weak positive correlation between blood THg and  $\delta^{15}\text{N}$  values is likely to reflect, at least in part, the biomagnification of MeHg within marine food webs, such that individuals foraging at higher trophic levels were exposed to greater levels of Hg. However, the positive relationships between blood THg and  $\delta^{13}\text{C}$  values were stronger than with  $\delta^{15}\text{N}$  values (Fig. 3A and Fig. 3B). Biomagnification of Hg has been demonstrated in the Scotia Sea ecosystem, from particulate organic matter through to higher trophic levels (Anderson et al., 2009a; Seco et al., 2021). Indeed, mean blood THg concentrations of several large seabird species in our study region are higher than those of brown skuas in this study; however, it is notable that those of southern 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 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 Island in 2001/02 were lower than in brown skuas from Bird Island in 2014/15 (Anderson et al., 2009a; this study). The relationship between  $\delta^{15}\text{N}$  and Hg contamination is often apparent when comparing values among different species within seabird communities (e.g., Blévin et al., 2013; Anderson et al., 2009a), but is observed less frequently within a single species, and was apparent across our study populations despite relatively little variation in  $\delta^{15}\text{N}$  values (range: 10.2 to 12.8 ‰). Alternatively, because there is a baseline  $^{15}\text{N}$  enrichment from the Antarctic towards the subtropics in the Southern Ocean (Cherel and Hobson, 2007; Jaeger et al., 2010), the relationship between Hg and  $\delta^{15}\text{N}$  may simply reflect the increasing

contamination with latitude (i.e., trophic variation may be obscured by spatial variation). However, changes in baseline  $\delta^{15}\text{N}$  are less pronounced south of the APF (Cherel and Hobson, 2007; Jaeger et al., 2010). Moreover, differences in mean blood  $\delta^{15}\text{N}$  values among populations were small (means at Bird Island are  $\sim 0.2$  ‰ higher than at Bahía Esperanza/Hope Bay; Table 1), and mean  $\delta^{15}\text{N}$  values were slightly lower at Signy Island than at Bahía Esperanza/Hope Bay (Table 1).

#### 4.3. Sex differences in Hg contamination

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

#### 4.4. Could Hg contamination have fitness implications?

Hg is a neurotoxin and endocrine disruptor (Tan et al., 2009), and contamination may impact various aspects of development, physiology and breeding behaviour in seabirds (Burger and Gochfeld, 1997; Evers et al., 2008; Tartu et al., 2013; Goutte et al., 2015). Hg contamination has also been associated with reduced breeding success (Goutte et al., 2014; Ackerman et al., 2016; Mills et al., 2020). The mean blood THg (dw) concentrations reported here are equivalent 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 and Bahía Esperanza/Hope Bay, respectively (assuming a 79% moisture content) (Eagles-Smith et al., 2008; Ackerman et al., 2016). These values suggest a low risk of MeHg toxicity (Ackerman et al., 2016). Indeed, mean blood THg concentrations in our study were far lower

than those of brown skuas at Kerguelen; however, blood THg concentrations at Bird Island and Signy Island exceeded those of south polar skuas *Stercorarius maccormicki* at Adélie Land (Goutte et al., 2014; Table 3). Goutte et al. (2014) found short-term associations between Hg contamination and breeding success in both skua species, yet long-term reproductive impacts were more severe in south polar skuas than brown skuas, despite their lower blood THg concentrations. Selenium (Se) offers a well-known protective effect against Hg toxicity when in molar excess (i.e., when Se:Hg > 1), and blood Se:Hg molar ratios of south polar skuas were three times lower than the brown skuas (Carravieri et al., 2017; Manceau et al., 2021). Hence, although it currently appears unlikely that there are fitness consequences of Hg contamination for brown skuas in our study region (though we did not test this directly), the role of Se requires evaluation. Populations of brown skuas at Signy Island and Bird Island increased rapidly from the late-1950s to early-1980s (by 3.3% and 3.6% pa, respectively), but then rates slowed until the 2010s (0.4% pa) and 2000s (0.9% pa), respectively (Phillips et al., 2004; Carneiro et al., 2016b). Rather than a consequence of Hg contamination, the slower population growth rates seem more likely to be related to changes in prey availability and increased density-dependent competition (Phillips et al., 2004; Carneiro et al., 2016b).

## 5. Conclusion

Our study provides important baseline data on Hg contamination of brown skuas in the Southern Ocean, and analyses the underlying drivers of variation in contamination. Given the relatively high levels of Hg contamination of brown skuas at Bird Island, which in the early 2000s was likely the most densely-populated colony in the world (132 nesting pairs per km<sup>2</sup> of suitable habitat) (Phillips et al., 2004), further monitoring is required, particularly as there is some evidence for increasing Hg contamination of other seabirds at South Georgia (Mills et al., 2020). Moreover, MeHg concentrations peak in low-oxygen subsurface waters, and in a warming world, oxygen minimum zones are expected to increase, hence potentially enhancing methylation of Hg and its bioavailability to marine predators (Blum et al., 2013). Further work should also examine the potential protective effects of Se on Hg toxicity in brown skuas, and the risks posed by other contaminants to these skua populations, especially given the incidence of plastic ingestion by this species at the Antarctic Peninsula and the potential role of plastics in contaminant transmission (Lavers and Bond, 2016; Ibañez et al., 2020).

### **Declaration of competing interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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### **CRedit authorship statement**

WFM: Conceptualization, Methodology, Investigation, Formal Analysis, Visualization, Writing - Original Draft. AEI: Conceptualization, Methodology, Investigation, Resources, Writing - Review & Editing. PB: Investigation, Writing - Review & Editing. APBC: Resources, Investigation, Writing - Review & Editing. SB: Supervision, Writing - Review & Editing. YC: Supervision, Writing - Review & Editing. RM-J: Supervision, Investigation, Writing - Review & Editing. RARM: Investigation, Writing - Review & Editing. DM: Supervision, Writing - Review & Editing. SCV: Supervision, Writing - Review & Editing. RAP: Conceptualization, Methodology, Investigation, Resources, Supervision, Writing - Review & Editing.

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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	$1.02 \pm 0.50$	$11.61 \pm 0.49$	$-23.26 \pm 0.26$
		11	Female	$0.87 \pm 0.40$	$11.25 \pm 0.53$	$-23.06 \pm 0.41$
		23	Both	$0.95 \pm 0.45$	$11.44 \pm 0.53$	$-23.17 \pm 0.35$
Signy Island, South Orkney Islands	2014	15	Male	$3.92 \pm 2.30$	$11.44 \pm 0.48$	$-22.29 \pm 0.52$
		13	Female	$2.85 \pm 2.23$	$11.24 \pm 0.65$	$-22.10 \pm 0.39$
		28	Both	$3.42 \pm 2.29$	$11.35 \pm 0.56$	$-22.20 \pm 0.47$
Bird Island, South Georgia	2015	14	Male	$4.86 \pm 1.12$	$11.81 \pm 0.51$	$-20.17 \pm 0.22$
		17	Female	$4.15 \pm 1.00$	$11.50 \pm 0.61$	$-20.31 \pm 0.29$
		31	Both	$4.47 \pm 1.10$	$11.64 \pm 0.58$	$-20.24 \pm 0.26$

**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;  $\Delta\text{AIC}_C$  is the change in the Akaike information criterion corrected for small sample sizes ( $\text{AIC}_C$ ) from the best-supported model;  $\omega_i$  is the Akaike weight; Exp. Dev. is the percentage explained deviance of the top model.

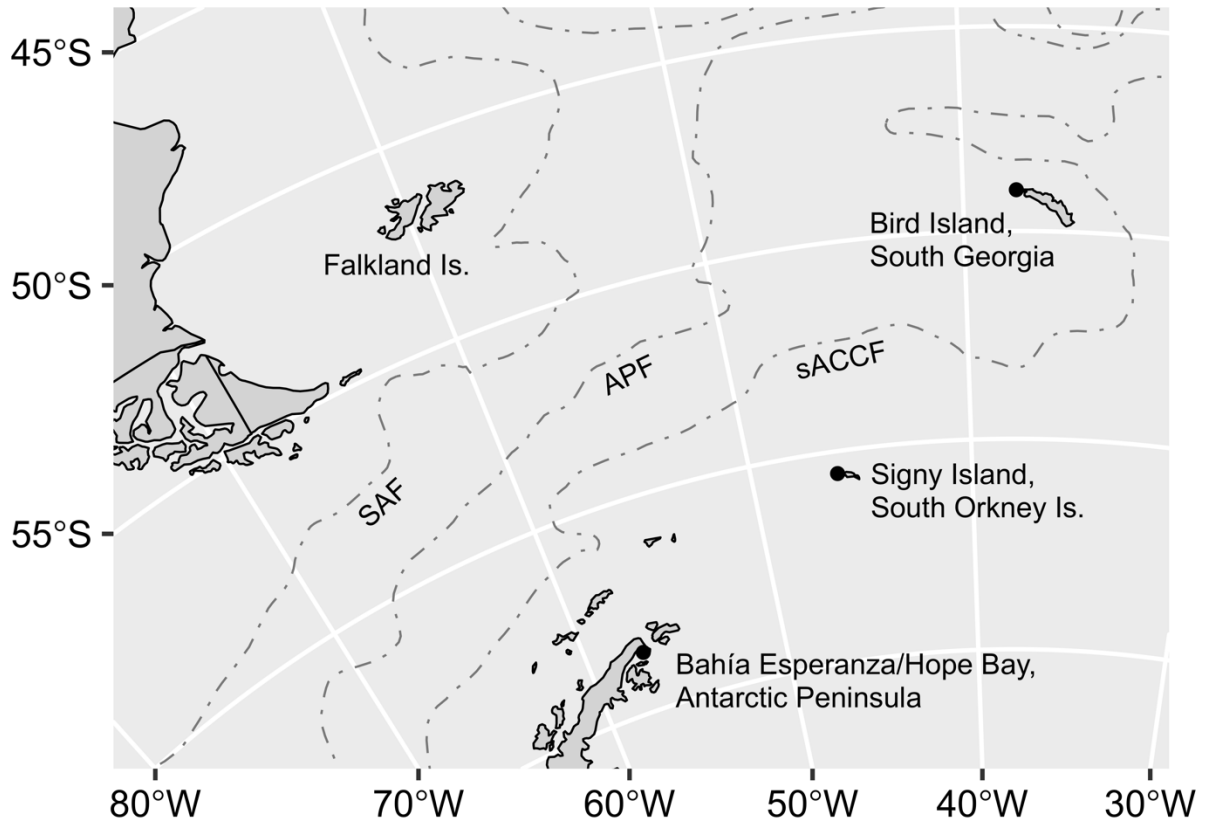
Models	k	$\Delta\text{AIC}_C$	$\omega_i^a$	Exp. Dev. (%)
<hr/> Maximal model: $\text{THg} \sim \text{Sex} + \text{Site} + \delta^{15}\text{N} + \text{Sex}:\delta^{15}\text{N} + \text{Site}:\text{Sex} + \text{Site}:\delta^{15}\text{N}$ <hr/>				
$\text{THg} \sim \text{Sex} + \text{Site} + \delta^{15}\text{N} + \text{Sex}:\delta^{15}\text{N}$	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 <sup>b</sup> : $\text{THg} \sim \text{Sex} + \delta^{13}\text{C} + \delta^{15}\text{N} + \text{Sex}:\delta^{13}\text{C} + \text{Sex}:\delta^{15}\text{N}$ <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	

<sup>a</sup>Weights across all models sum to 1.00 (not all shown).

<sup>b</sup>We did not consider an interaction between blood  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values.

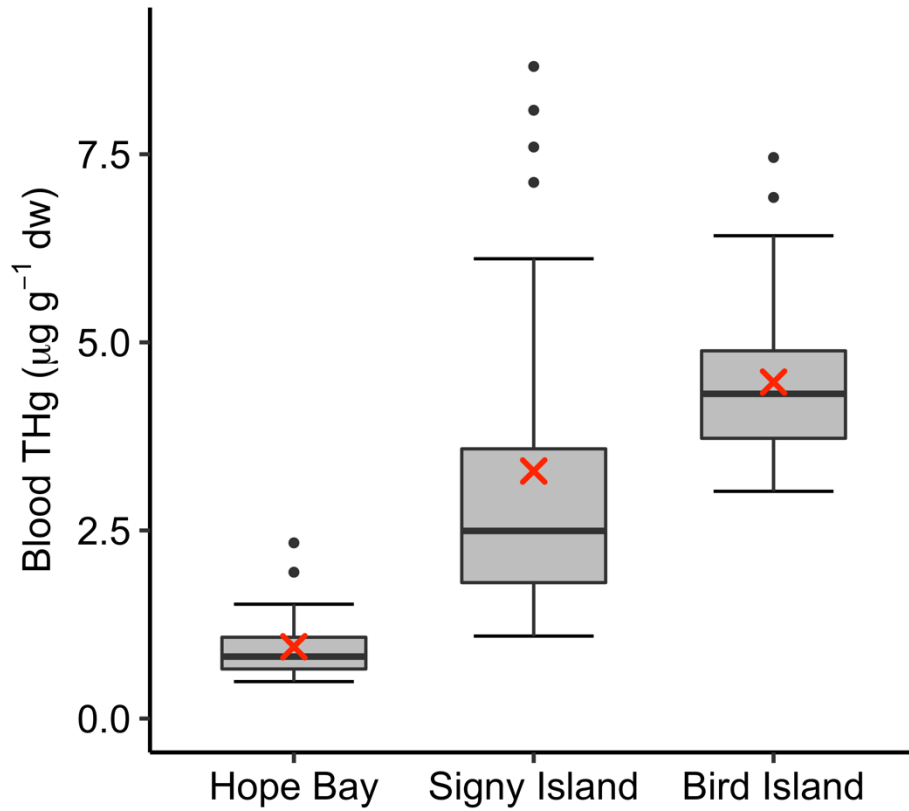
**Table 3.** Review of published blood total mercury (THg) concentrations ( $\mu\text{g g}^{-1}$  dw) in adults and chicks of large skua species in the Northern and Southern Hemispheres. Values are means  $\pm$  SDs.

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

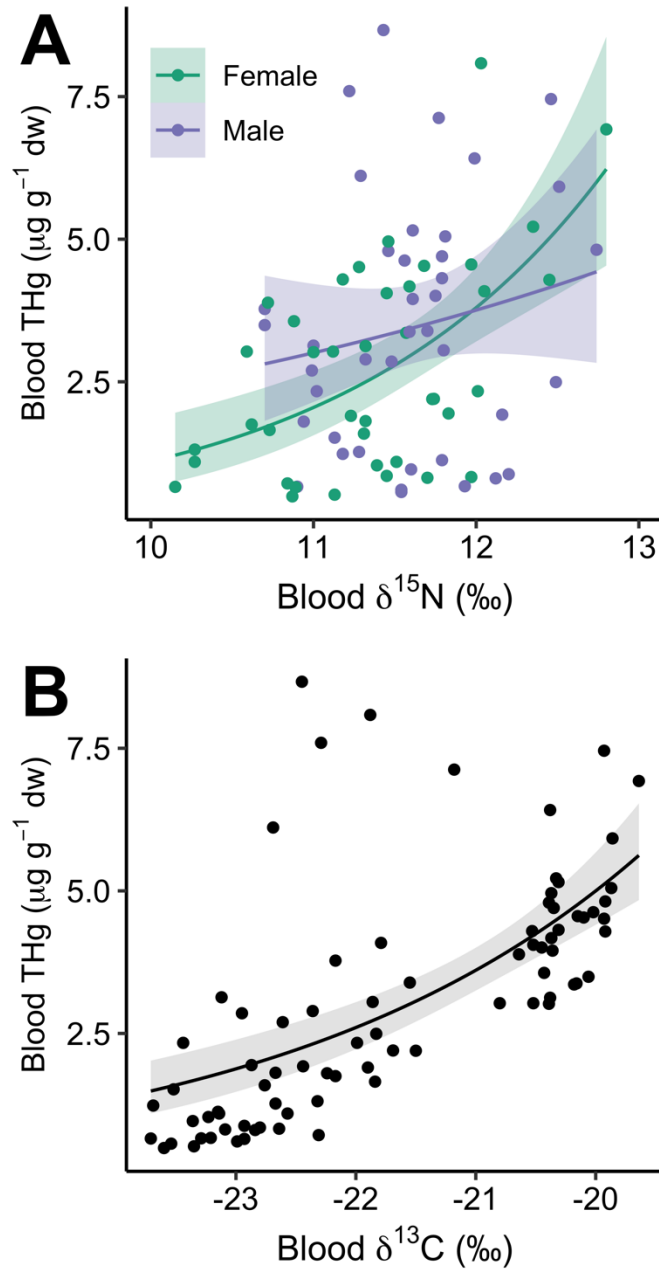


**Figure 1.** Location of the three study sites in relation to the Sub-Antarctic Front (SAF), the Antarctic Polar Front (APF) and the southern Antarctic Circumpolar Current Front (sACCF). The study sites are: (i) Bahía Esperanza/Hope Bay, Antarctic Peninsula ( $63^{\circ}24'S$ ,  $57^{\circ}01'W$ ); (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 ( $54^{\circ}00'\text{S}$ ,  $38^{\circ}03'\text{W}$ ).