



## Mercury biomagnification in a Southern Ocean food web<sup>☆</sup>

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### ABSTRACT

Biomagnification of mercury (Hg) in the Scotia Sea food web of the Southern Ocean was examined using the stable isotope ratios of nitrogen ( $\delta^{15}\text{N}$ ) and carbon ( $\delta^{13}\text{C}$ ) as proxies for trophic level and feeding habitat, respectively. Total Hg and stable isotopes were measured in samples of particulate organic matter (POM), zooplankton, squid, myctophid fish, notothenioid fish and seabird tissues collected in two years (austral summers 2007/08 and 2016/17). Overall, there was extensive overlap in  $\delta^{13}\text{C}$  values across taxonomic groups suggesting similarities in habitats, with the exception of the seabirds, which showed some differences, possibly due to the type of tissue analysed (feathers instead of muscle).  $\delta^{15}\text{N}$  showed increasing enrichment across groups in the order POM to zooplankton to squid to myctophid fish to notothenioid fish to seabirds. There were significant differences in  $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$  values among species within taxonomic groups, reflecting inter-specific variation in diet. Hg concentrations increased with trophic level, with the lowest values in POM ( $0.0005 \pm 0.0002 \mu\text{g g}^{-1} \text{dw}$ ) and highest values in seabirds ( $3.88 \pm 2.41 \mu\text{g g}^{-1}$  in chicks of brown skuas *Stercorarius antarcticus*). Hg concentrations tended to be lower in 2016/17 than in 2007/08 for mid-trophic level species (squid and fish), but the opposite was found for top predators (i.e. seabirds), which had higher levels in the 2016/17 samples. This may reflect an interannual shift in the Scotia Sea marine food web, caused by the reduced availability of a key prey species, Antarctic krill *Euphausia superba*. In 2016/17, seabirds would have been forced to feed on higher trophic-level prey, such as myctophids, that have higher Hg burdens. These results suggest that changes in the food web are likely to affect the pathway of mercury to Southern Ocean top predators.

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## 1. Introduction

Antarctica and the Southern Ocean comprise a globally unique

ecosystem characterised by cold environmental conditions. Due to its isolation and often-inhospitable weather, much of the region has remained relatively untouched by direct human activity, such that it is often considered by the scientific community as a natural laboratory (Walton, 2013). Given the absence of any local manufacturing industry, anthropogenic mercury (Hg) emissions from within Antarctica are negligible. Nevertheless, there is long-range dispersal of Hg from regions outside Antarctica, and

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relatively high concentrations can be found in Southern Ocean waters (Cossa et al., 2011). The global distillation process (Wania and Mackay, 1996) leads to atmospheric transportation of a range of volatile and semi-volatile pollutants in the form of vapour. Hg is transported by atmospheric currents in the form of  $\text{Hg}^0$  to the polar regions where it condenses and precipitates in rain or snow (O'Driscoll et al., 2005). Indeed, Antarctica can be considered a gigantic fridge where atmospherically transported pollutants are stored in the ice fields (Eisele et al., 2008), becoming bioavailable when glaciers or icebergs melt (Mastromonaco et al., 2017). Moreover, recent studies have shown that vegetation in the polar environments can play an important role in the uptake of  $\text{Hg}^0$ . Plus, Atmospheric Mercury Depletion Events (AMDE) occurring during springtime at polar sunrise (Ebinghaus et al., 2002) are known to increase the deposition fluxes of atmospheric Hg (Brooks et al., 2008; Elizalde, 2017), and katabatic winds can redistribute Hg within Antarctica, carrying it from the high Antarctic plateau towards coastal areas (Bargagli, 2016; Bromwich, 1989).

Among pollutants, Hg is one of the most toxic elements, particularly its organic form (methyl-Hg,  $[\text{CH}_3\text{Hg}]^+$ ) (Clarkson, 1992). Due to its high affinity for proteins and hence retention within tissues (Bloom, 1992), Hg is highly bioaccumulative in organisms over the course of their lives. It also biomagnifies along food webs from plankton up to top predators (Ackerman et al., 2014; Coelho et al., 2013; Dehn et al., 2006). Methyl-Hg is assimilated more efficiently by organisms than inorganic Hg, and is accumulated rather than excreted (Monteiro et al., 1996). Hg concentrations are therefore higher in the upper trophic levels of food webs, especially those in aquatic systems which tend to be highly size-structured (Heneghan et al., 2019), and may become very toxic for large, long-lived top predators (Goutte et al., 2014; Tartu et al., 2014; Tavares et al., 2013). Indeed, some Southern Ocean predators have particularly high Hg concentrations. For example, wandering albatross, *Diomedea exulans*, which can live to over 50 years (Lecomte et al., 2010) have amongst the highest reported feather Hg concentrations (up to  $73.42 \mu\text{g g}^{-1}$ ) of any seabird, reflecting a high degree of exposure to this metal (Anderson et al., 2009; Cherel et al., 2018; Tavares et al., 2013; Thompson et al., 1998).

Indices of the enrichment of stable isotopes of nitrogen ( $\delta^{15}\text{N}$ ) and carbon ( $\delta^{13}\text{C}$ ) provide proxies for the trophic level and carbon source (habitat) of consumers, respectively (Cherel and Hobson, 2007; Stowasser et al., 2012).  $^{15}\text{N}$  becomes enriched in tissues in a consistent way in food webs, on average by  $+3.4 \text{‰}$  per trophic level (Minagawa and Wada, 1984).  $^{13}\text{C}$  is enriched at a lower rate (on average by  $+1 \text{‰}$  per trophic level) but gives a useful indicator of feeding habitat; this indicates, for example, the relative contributions of feeding inshore versus offshore, in shallow water versus deep water, or in particular water masses given the broad latitudinal gradients in the Southern Ocean (Hobson et al., 1994). Although some studies have reported Hg concentrations both in low trophic level organisms and top predators in the Southern Ocean (e.g. Anderson et al., 2009), none have evaluated Hg biomagnification across multiple levels in the food web, from particulate organic matter (POM) to apex predators, using  $\delta^{15}\text{N}$  to determine trophic position.

Antarctic krill (*Euphausia superba*; hereafter krill) is a key component of the food web in the Southern Ocean (Murphy et al., 2007), and can be the main trophic link between primary producers and predators (Everson, 2000; Xavier and Peck, 2015; Xavier et al., 2018). However, recent studies have shown that there are alternative trophic pathways in periods and regions of low Antarctic krill abundance. These include, for example, copepods to mesopelagic fish to high predators (Ballerini et al., 2014; Murphy et al., 2007, 2013; Saunders et al., 2019). These alternative pathways are unlikely to support the same biomass of predators given the greater

number of steps in the food chain over which energy can be lost (Barnes et al., 2010). Decreases in the reproductive performance of top predators, including seabirds and pinnipeds, in low-krill years in the Scotia Sea region suggests that these alternative pathways cannot entirely replace those involving krill in maintaining very large predator populations (Croxall et al., 1999; Xavier et al., 2003, 2017).

In the present study, we measured total Hg concentrations,  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values in a suite of species from different trophic levels to elucidate Hg biomagnification in components of the food web in the Scotia Sea, one of the most productive regions of the Southern Ocean (Atkinson et al., 2001). POM and tissues from key functional organisms from multiple trophic levels were analysed, including zooplankton, squid, mesopelagic fish (myctophids), necto-benthic fish (notothenioids), and seabirds. The field sampling was undertaken in two austral summers, 9 years apart (December 2007 to February 2008 and December 2016 to January 2017). The main objectives were to 1) describe Hg dynamics in the Scotia Sea food web, 2) evaluate Hg biomagnification-rates, and, 3) evaluate possible differences in the Hg pathway from POM to top predators in different sampling years.

## 2. Materials and methods

### 2.1. Field sampling

Samples were collected during oceanographic research cruises on board the RRS *James Clark Ross* around the islands of South Georgia ( $54^\circ 17' \text{S}$ ,  $36^\circ 30' \text{W}$ ) during the austral summers of 2007/08 and 2016/17 (cruises JR177 and JR16003 respectively). Background concentrations of Hg in POM were determined from water samples collected in Niskin bottles deployed on a CTD (conductivity, temperature, depth) rosette fired at the depth of the *chlorophyll a* maximum (which ranged from 30 to 76 m) and at 500 m. The depths were chosen to enable comparison of the likely highest concentrations of POM with the lower values expected below the euphotic zone. POM was obtained by vacuum-filtering 5 L of water through glass fibre filters (GF/F Whatman, 47 mm) during JR16003. Zooplankton, squid and myctophid fish species were collected using either an 8 or 25  $\text{m}^2$  mouth-opening Rectangular Midwater Trawl [RMT8 - mesh size reducing from 4.5 mm to 2.5 mm in the cod end to collect zooplankton; RMT25 - mesh size reducing from 8 mm to 4.5 mm in the cod end to collect mesopelagic fish and squid (Roe and Shale, 1979)]. Both systems incorporated two separate nets that could be opened and closed remotely on command at different depths. Myctophid fish and squid were identified using appropriate taxonomic keys (Gon and Heemstra, 1990; Hulley, 1981; Nesis, 1987; Xavier and Cherel, 2009). Sub-samples were then frozen individually in separate plastic bags at  $-20 \text{ °C}$  for later laboratory analyses. Zooplankton species were identified following Boltovskoy (1999). Sub-samples of these species were either preserved in plastic bags at  $-20 \text{ °C}$  (JR16003) or in glass vials in 70% ethanol (JR177).

Notothenioid fish were also obtained from South Georgia waters in the austral summer of 2016/17. They were caught by the Fishing Vessel (FV) *Sil* during research survey SG17. Samples were obtained from bottom trawls using a FP120 trawl net with a standard steel bobbin rig. Whenever possible, samples were identified on board but, in some cases, identification was not possible at sea and was performed later at the laboratory. Individuals were frozen at sea at  $-20 \text{ °C}$  for later laboratory processing.

Feathers from seabird chicks were collected at Bird Island ( $54^\circ 00' \text{S}$ ,  $38^\circ 03' \text{W}$ ), South Georgia, in austral summers 2007/08 and 2016/17. Chicks were sampled rather than adults because Hg and stable isotope ratios in their tissues reflect those of food

consumed during the chick-rearing period (Blévin et al., 2013; Moreno et al., 2016). Furthermore, all of these feathers are grown by chicks over the same period, so Hg concentrations and stable isotope ratios reflect similar time periods and can be compared directly (Carravieri et al., 2014a).

We decided to analyse feathers in the seabirds for a number of reasons: (1) it is a less invasive and stressful sampling procedure than collection of blood, and it would have been inappropriate to sacrifice these species just for Hg analysis, particularly as most are protected, (2) feathers have been widely used as a means to monitor Hg in seabirds, it is therefore easier to compare results with those of other studies, (3) several previous works have compared the Hg in feathers with those in muscle tissue or whole prey (e.g., Goutner and Furness, 1997; Bargagli et al., 1998; Monteiro et al., 1998; Cristol et al., 2008; Fort et al., 2016; Abeyasinghe et al., 2017), (4) in most seabird species, feathers can hold up to 90% of the total Hg body burden (Braune and Gaskin, 1987; Atwell et al., 1998).

## 2.2. Laboratory procedures

POM filters were digested with HNO<sub>3</sub> 4 M for determination of Hg concentrations (for details, see Pato et al., 2010). Analyses were performed at the University of Aveiro by cold-vapour atomic fluorescence spectrometry (CV-AFS) using a PSA model Merlin 10.023 equipped with a PSA model 10.003 detector, with tin chloride as a reducing agent (2% in 10% HCl) following Pato et al. (2010). The limit of quantification of this technique was 0.02 µg L<sup>-1</sup>.

Zooplankton were analysed as whole individuals. Specimens that had been preserved in 70% ethanol were first dried for 72 h at ambient temperature to remove ethanol, and then freeze-dried for 48 h (Fort et al., 2014). Samples of squid and fish muscle tissue (freeze-dried for > 24 h) were chosen for analysis since muscle is the most important tissue in terms of transfer of Hg to predators (Bustamante et al., 2006; Cipro et al., 2018). Feathers were first cleaned to remove surface contaminants using a 2:1 chloroform:methanol solution followed by two methanol rinses, and then oven dried for 48 h at 50 °C.

Dried individual zooplankton, muscle samples and feathers were homogenized to powder and analysed for total Hg by thermal decomposition atomic absorption spectrometry with gold amalgamation, using a LECO AMA-254 (Advanced Mercury Analyzer) at the University of Aveiro, following Coelho et al. (2008). Analytical quality control was performed using certified reference materials (CRMs): for zooplankton we used TORT-2 and TORT-3 with recoveries of 87 ± 3% and 90 ± 8%, respectively; for squid we used NIST 2976, ERM-CE278K and TORT-3 with recoveries of 85 ± 7%, 92 ± 5% and 93 ± 8%, respectively; for myctophids we used DORM-4 and ERM-BB422 with recoveries of 96 ± 13% and 100 ± 4%, respectively; for notothenioid fish we used ERM-BB422 with recovery of 98 ± 7%, and for seabirds we used TORT-3 with recovery of 99 ± 3%. Sample analyses were repeated in duplicate or triplicate until the relative standard deviation was <10% for multiple aliquots. The limit of detection for this analytical method was 0.01 ng of absolute Hg.

As lipid is enriched in <sup>13</sup>C relative to other tissue components (DeNiro and Epstein, 1977), lipids were extracted from fish muscle. An aliquot of approximately 10 mg of fine muscle-tissue powder was agitated with 4 ml of cyclohexane for 1 h. Next, the sample was centrifuged for 5 min at 4000 g, and the supernatant containing lipids was discarded. Due to small masses and low sample numbers it was not possible to remove lipids from zooplankton, nor to pool samples, so stable isotope ratios were corrected according to Post et al. (2007). As squid muscle and bird feathers have low lipid content, no delipidation was applied. Low lipid content was

checked by considering C/N mass ratios, which were always below 4.0 (squid – 3.91 and seabirds – 3.46).

Approximately 0.4 mg of dry subsample homogenates were weighed out into tin cups. A continuous flow mass spectrometer (Thermo Scientific Delta V Advantage) was coupled to an elemental analyzer (Thermo Scientific Flash EA 1112), at either the LIENSs or MAREFOZ laboratories, to measure δ<sup>13</sup>C and δ<sup>15</sup>N values. Stable isotope ratios are expressed using standard δ notation relative to carbonate Vienna PeeDee Belemnite and atmospheric nitrogen. The internal laboratory standard is acetanilide. Observed analytical errors were <0.10‰ for both δ<sup>13</sup>C and δ<sup>15</sup>N values at both facilities.

## 2.3. Statistical analysis

All analyses were performed using R software v. 3.4.2 (R Core Team, 2013). Distributions of Hg concentrations and δ<sup>13</sup>C and δ<sup>15</sup>N values within samples were tested for normality using Shapiro-Wilk normality test, and homogeneity of variance was tested using Bartlett's test. Wilcoxon rank and Kruskal–Wallis tests were used to compare Hg, δ<sup>13</sup>C and δ<sup>15</sup>N values among trophic groups (zooplankton, squid, myctophid fish, notothenioid fish, and seabirds) and species, followed by a Dunn's multiple comparisons test. T-test or Mann-Whitney were used to compare Hg, δ<sup>13</sup>C and δ<sup>15</sup>N values between 2007/08 and 2016/17. Linear regressions were examined between Log<sub>10</sub>Hg and δ<sup>15</sup>N values as a tool to evaluate the trophic magnification slope (TMS) (Lavoie et al., 2013). All values are presented as means ± SD. The significance level for statistical analyses was α = 0.05.

## 3. Results

### 3.1. δ<sup>13</sup>C as a proxy of habitat

δ<sup>13</sup>C values for samples collected in 2007/08 and 2016/17 are given in Tables 1 and 2 respectively. Among whole zooplankton, δ<sup>13</sup>C values ranged from –25.64 ‰ in krill (*Euphausia superba*) to –20.48 ‰ in the amphipod *Parandania boeckii*. For secondary consumers (myctophid fish and squid), δ<sup>13</sup>C values in muscle tissue ranged from –25.67 ‰ (*Electrona antarctica*) to –20.15 ‰ (*Slosarczykovia circumantarctica*), while for notothenioid fish, it was from –23.80‰ in *Champscephalus gunnari* to –20.60 ‰ in *Dissostichus eleginoides*. Values of δ<sup>13</sup>C in seabird feathers tended to be higher (i.e. more enriched in <sup>13</sup>C), ranging from –22.82 ‰ in Antarctic prions (*Pachyptila desolata*) to –15.24 ‰ in brown skuas (*Stercorarius antarcticus*) (Table 1). There were only significant differences in δ<sup>13</sup>C values between seabirds and the other trophic groups within each sampling year (Kruskal-Wallis test, H = 50.23, p < 0.0001, in 2007/08; H = 63.69, p < 0.0001 in 2016/17). There were no differences between the other groups.

Within the seabird group, brown skuas had significantly higher δ<sup>13</sup>C values than northern giant petrels (*Macronectes halli*) in 2007/08 (Kruskal-Wallis test, H = 15.932, p = 0.007; Dunn's multiple comparisons test, p = 0.0028; Table 1). In 2016/17, significant differences among seabirds were also detected (Kruskal-Wallis test, H = 22.552, p < 0.0001; Table 2) and these were mainly driven by the higher value in brown skuas compared with Antarctic prions (Dunn's multiple comparisons test, p = 0.0013), and blue petrels *Halobaena caerulea* (Dunn's multiple comparisons test, p = 0.0144).

Within the notothenioid fish (Kruskal-Wallis test, H = 24.794, p = 0.0002; Table 2), *N. gibberifrons* had significant higher δ<sup>13</sup>C than *C. gunnari* (Dunn's multiple comparisons test, p = 0.0002) and *P. guntheri* (Dunn's multiple comparisons test, p = 0.00013). No significant differences in δ<sup>13</sup>C were observed between any myctophid fish in 2007/08 (Kruskal-Wallis test, H = 16.141, p = 0.061; Table 1) whereas, in 2016/17, δ<sup>13</sup>C values were significantly lower in

**Table 1**

Mercury (Hg) concentration,  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values of different species sampled on land or caught in the waters around South Georgia in the 2007/08 austral summer (mean  $\pm$  standard deviation). The superscript numbers (<sup>1</sup>) and letters (<sup>a</sup>) denotes statistical significance from within the sampling year and comparison between years, respectively.

Species	n	Hg ( $\mu\text{g g}^{-1}$ dw)	$\delta^{15}\text{N}$ (‰)	$\delta^{13}\text{C}$ (‰)
<b>Zooplankton</b>				
<i>Parandania boeckii</i>	15	0.02 $\pm$ 0.01 <sup>1,a</sup>	8.34 $\pm$ 0.42 <sup>1,a</sup>	-21.90 $\pm$ 1.70 <sup>1,a</sup>
<i>Euphausia triacantha</i>	20	0.03 $\pm$ 0.01 <sup>1,a</sup>	6.71 $\pm$ 0.55 <sup>1,2,a</sup>	-22.15 $\pm$ 1.47 <sup>1,a</sup>
<i>Gigantocypris</i> sp.	15	0.03 $\pm$ 0.01 <sup>1,a</sup>	8.81 $\pm$ 0.98 <sup>1,2,a</sup>	-22.27 $\pm$ 0.41 <sup>1,a</sup>
<i>Salpa thompsoni</i>	10	0.03 $\pm$ 0.01 <sup>1</sup>	4.40 $\pm$ 0.22 <sup>1,2</sup>	-23.75 $\pm$ 1.32 <sup>1</sup>
<i>Tomopteris</i> sp.	6	0.03 $\pm$ 0.01 <sup>1</sup>	7.25 $\pm$ 0.73 <sup>1,2</sup>	-22.64 $\pm$ 0.65 <sup>1</sup>
<i>Euphausia superba</i>	20	0.04 $\pm$ 0.02 <sup>1,a</sup>	3.47 $\pm$ 2.45 <sup>2,a</sup>	-23.17 $\pm$ 2.86 <sup>1,a</sup>
<i>Themisto gaudichaudii</i>	20	0.04 $\pm$ 0.02 <sup>1,a</sup>	5.86 $\pm$ 0.56 <sup>1,2,a</sup>	-24.10 $\pm$ 0.45 <sup>1,a</sup>
<i>Thysanoessa</i> sp.	20	0.05 $\pm$ 0.01 <sup>1,a</sup>	6.94 $\pm$ 0.32 <sup>1,2,a</sup>	-23.99 $\pm$ 0.71 <sup>1,a</sup>
<i>Sagitta</i> sp.	3	0.06 $\pm$ 0.01 <sup>1</sup>	8.21 $\pm$ 0.84 <sup>1,2</sup>	-22.97 $\pm$ 1.03 <sup>1</sup>
<b>Squid</b>				
<i>Slosarczykovia circumantarctica</i>	5	0.02 $\pm$ 0.01 <sup>1,a</sup>	6.72 $\pm$ 0.25 <sup>1,a</sup>	-21.64 $\pm$ 1.88 <sup>1,b</sup>
<i>Galiteuthis glacialis</i>	5	0.09 $\pm$ 0.01 <sup>1,a</sup>	7.66 $\pm$ 1.49 <sup>1,a</sup>	-23.58 $\pm$ 1.05 <sup>1,a</sup>
<b>Myctophid fish</b>				
<i>Krefflichthys anderssoni</i>	5	0.04 $\pm$ 0.01 <sup>2,a</sup>	8.21 $\pm$ 0.95 <sup>1,a</sup>	-20.96 $\pm$ 0.19 <sup>1,a</sup>
<i>Protomyctophum bolini</i>	5	0.09 $\pm$ 0.01 <sup>2,a</sup>	8.87 $\pm$ 0.88 <sup>1,a</sup>	-21.82 $\pm$ 2.13 <sup>1,a</sup>
<i>Electrona carlsbergi</i>	5	0.14 $\pm$ 0.02 <sup>2,3</sup>	8.31 $\pm$ 1.12 <sup>1</sup>	-20.91 $\pm$ 0.83 <sup>1</sup>
<i>Gymnoscopelus braueri</i>	5	0.16 $\pm$ 0.03 <sup>2,3,a</sup>	9.77 $\pm$ 0.56 <sup>1,a</sup>	-23.63 $\pm$ 0.44 <sup>1,a</sup>
<i>Gymnoscopelus opisthopterus</i>	5	0.16 $\pm$ 0.06 <sup>2,3</sup>	10.81 $\pm$ 0.15 <sup>1</sup>	-23.12 $\pm$ 0.96 <sup>1</sup>
<i>Electrona antarctica</i>	5	0.18 $\pm$ 0.09 <sup>2,3,b</sup>	8.64 $\pm$ 0.96 <sup>1,a</sup>	-24.91 $\pm$ 0.66 <sup>1,a</sup>
<i>Gymnoscopelus nicholsi</i>	5	0.29 $\pm$ 0.12 <sup>3,a</sup>	8.70 $\pm$ 0.94 <sup>1,a</sup>	-22.00 $\pm$ 1.14 <sup>1,a</sup>
<b>Seabirds</b>				
<i>Macronectes halli</i>	5	0.47 $\pm$ 0.33 <sup>4,b</sup>	13.07 $\pm$ 0.56 <sup>3,4,b</sup>	-18.68 $\pm$ 0.87 <sup>2,a</sup>
<i>Macronectes giganteus</i>	5	0.85 $\pm$ 0.12 <sup>5,b</sup>	11.80 $\pm$ 0.33 <sup>3,a</sup>	-18.23 $\pm$ 0.66 <sup>2,3,b</sup>
<i>Thalassarche melanophris</i>	5	1.18 $\pm$ 0.46 <sup>5,a</sup>	11.83 $\pm$ 0.19 <sup>3,a</sup>	-17.30 $\pm$ 0.93 <sup>2,3,b</sup>
<i>Thalassarche chrysostoma</i>	5	1.25 $\pm$ 0.45 <sup>5,a</sup>	12.06 $\pm$ 0.56 <sup>3,a</sup>	-18.22 $\pm$ 1.47 <sup>2,3,b</sup>
<i>Stercorarius antarcticus</i>	5	2.44 $\pm$ 1.08 <sup>6,a</sup>	12.63 $\pm$ 0.54 <sup>3,4,a</sup>	-15.59 $\pm$ 0.32 <sup>3,b</sup>
<i>Diomedea exulans</i>	5	3.28 $\pm$ 0.63 <sup>6,a</sup>	14.83 $\pm$ 0.61 <sup>4,a</sup>	-18.29 $\pm$ 0.42 <sup>2,3,b</sup>

**Table 2**

Mercury (Hg) concentration,  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values of different species sampled on land or caught in the waters around South Georgia in the 2016/17 austral summer (mean  $\pm$  standard deviation). The superscript numbers (<sup>1</sup>) and letters (<sup>a</sup>) denotes statistical significance from within the sampling year and comparison between years, respectively.

Species	n	Hg ( $\mu\text{g g}^{-1}$ dw)	$\delta^{15}\text{N}$ (‰)	$\delta^{13}\text{C}$ (‰)
POM	12	0.0005 $\pm$ 0.0002	n.a.	n.a.
<b>Zooplankton</b>				
<i>Euphausia superba</i>	30	0.01 $\pm$ 0.003 <sup>1,a</sup>	3.38 $\pm$ 0.38 <sup>1,a</sup>	-24.98 $\pm$ 0.66 <sup>1,a</sup>
<i>Euphausia vallentini</i>	30	0.01 $\pm$ 0.03 <sup>2</sup>	3.00 $\pm$ 0.78 <sup>1</sup>	-21.60 $\pm$ 0.18 <sup>1</sup>
<i>Thysanoessa</i> sp.	40	0.02 $\pm$ 0.01 <sup>2,a</sup>	5.62 $\pm$ 0.29 <sup>2,a</sup>	-23.99 $\pm$ 0.64 <sup>1,a</sup>
<i>Euphausia triacantha</i>	30	0.02 $\pm$ 0.03 <sup>2,a</sup>	7.08 $\pm$ 0.62 <sup>2,a</sup>	-22.72 $\pm$ 0.17 <sup>1,a</sup>
<i>Euphausia frigida</i>	35	0.05 $\pm$ 0.01 <sup>2</sup>	5.94 $\pm$ 0.04 <sup>2</sup>	-21.16 $\pm$ 0.48 <sup>1</sup>
<i>Themisto gaudichaudii</i>	40	0.06 $\pm$ 0.02 <sup>2,a</sup>	5.61 $\pm$ 0.79 <sup>2,a</sup>	-22.46 $\pm$ 0.52 <sup>1,a</sup>
<i>Gigantocypris</i> sp.	20	0.07 $\pm$ 0.01 <sup>2,a</sup>	7.07 $\pm$ 1.09 <sup>2,a</sup>	-24.71 $\pm$ 0.35 <sup>1,a</sup>
<i>Euphausia spinifera</i>	20	0.07 $\pm$ 0.02 <sup>2</sup>	5.05 $\pm$ 0.66 <sup>2</sup>	-22.79 $\pm$ 0.09 <sup>1</sup>
<i>Parandania boeckii</i>	30	0.12 $\pm$ 0.03 <sup>2,a</sup>	7.55 $\pm$ 0.28 <sup>2,a</sup>	-21.82 $\pm$ 1.90 <sup>1,a</sup>
<b>Squid</b>				
<i>Slosarczykovia circumantarctica</i>	5	0.01 $\pm$ 0.01 <sup>2,a</sup>	6.23 $\pm$ 0.78 <sup>2,a</sup>	-23.13 $\pm$ 0.43 <sup>1,b</sup>
<i>Galiteuthis glacialis</i>	5	0.02 $\pm$ 0.01 <sup>2,a</sup>	7.40 $\pm$ 0.09 <sup>2,a</sup>	-24.68 $\pm$ 0.49 <sup>1,a</sup>
<b>Myctophid fish</b>				
<i>Krefflichthys anderssoni</i>	5	0.05 $\pm$ 0.01 <sup>2,a</sup>	8.48 $\pm$ 0.50 <sup>2,a</sup>	-23.02 $\pm$ 0.10 <sup>1,a</sup>
<i>Protomyctophum bolini</i>	5	0.10 $\pm$ 0.03 <sup>2,a</sup>	7.98 $\pm$ 0.75 <sup>2,a</sup>	-23.43 $\pm$ 0.70 <sup>1,a</sup>
<i>Gymnoscopelus braueri</i>	5	0.12 $\pm$ 0.06 <sup>2,3,a</sup>	9.58 $\pm$ 0.97 <sup>2,a</sup>	-24.37 $\pm$ 0.61 <sup>1,a</sup>
<i>Electrona antarctica</i>	5	0.12 $\pm$ 0.07 <sup>2,3,b</sup>	7.41 $\pm$ 0.72 <sup>2,a</sup>	-24.54 $\pm$ 0.34 <sup>1,a</sup>
<i>Gymnoscopelus nicholsi</i>	5	0.30 $\pm$ 0.17 <sup>3,a</sup>	9.73 $\pm$ 0.03 <sup>2,a</sup>	-20.74 $\pm$ 0.34 <sup>1,a</sup>
<b>Notothenioid fish</b>				
<i>Champscephalus gunnari</i>	11	0.02 $\pm$ 0.01 <sup>2</sup>	9.18 $\pm$ 0.56 <sup>2</sup>	-22.95 $\pm$ 0.44 <sup>1</sup>
<i>Patagonotothen guntheri</i>	5	0.10 $\pm$ 0.03 <sup>2,3</sup>	8.34 $\pm$ 0.56 <sup>2</sup>	-23.43 $\pm$ 0.32 <sup>1,2</sup>
<i>Chaenocephalus aceratus</i>	5	0.11 $\pm$ 0.02 <sup>2,3</sup>	10.79 $\pm$ 1.12 <sup>2,3</sup>	-21.66 $\pm$ 0.50 <sup>1,2</sup>
<i>Notothenia gibberifrons</i>	8	0.18 $\pm$ 0.07 <sup>3</sup>	11.30 $\pm$ 0.54 <sup>3</sup>	-19.93 $\pm$ 0.78 <sup>2</sup>
<i>Notothenia rossii</i>	8	0.18 $\pm$ 0.08 <sup>3</sup>	10.78 $\pm$ 1.67 <sup>2,3</sup>	-21.39 $\pm$ 0.58 <sup>1,2</sup>
<i>Dissostichus eleginoides</i>	5	0.20 $\pm$ 0.06 <sup>3</sup>	11.86 $\pm$ 0.75 <sup>3</sup>	-21.76 $\pm$ 0.77 <sup>1</sup>
<b>Seabirds</b>				
<i>Pachyptila desolata</i>	5	0.22 $\pm$ 0.14 <sup>3</sup>	8.59 $\pm$ 0.78 <sup>2</sup>	-21.58 $\pm$ 0.73 <sup>3</sup>
<i>Halobaena caerulea</i>	5	0.62 $\pm$ 0.23 <sup>3</sup>	9.25 $\pm$ 0.50 <sup>2,3</sup>	-21.14 $\pm$ 0.67 <sup>3</sup>
<i>Thalassarche chrysostoma</i>	5	1.43 $\pm$ 0.50 <sup>4,a</sup>	11.78 $\pm$ 0.67 <sup>3,a</sup>	-19.91 $\pm$ 0.40 <sup>3,4,b</sup>
<i>Thalassarche melanophris</i>	5	1.51 $\pm$ 0.46 <sup>4,a</sup>	11.52 $\pm$ 0.35 <sup>2,3,a</sup>	-20.32 $\pm$ 0.63 <sup>3,4,b</sup>
<i>Macronectes giganteus</i>	5	1.68 $\pm$ 0.27 <sup>4,b</sup>	11.52 $\pm$ 0.41 <sup>2,3,a</sup>	-20.46 $\pm$ 0.38 <sup>3,4,b</sup>
<i>Macronectes halli</i>	5	2.05 $\pm$ 0.80 <sup>4,b</sup>	11.58 $\pm$ 0.65 <sup>2,3,b</sup>	-19.69 $\pm$ 1.12 <sup>3,4,a</sup>
<i>Stercorarius antarcticus</i>	5	3.88 $\pm$ 2.41 <sup>5,a</sup>	11.88 $\pm$ 1.13 <sup>3,a</sup>	-18.79 $\pm$ 0.90 <sup>4,b</sup>

*E. antarctica* than in *Gymnoscopelus nicholsi* (Kruskal-Wallis test,  $H = 16.751$ ,  $p = 0.0049$ ; Dunn's multiple comparisons test,  $p = 0.0115$ ; Table 2).

$\delta^{13}\text{C}$  values were not significantly different between sampling years either in squid (Wilcoxon rank test,  $W = 3$ ,  $p = 0.25$ , in 2007/08;  $W = 0$ ,  $p = 0.20$  in 2017) or zooplankton (Kruskal-Wallis test,  $H = 5.1538$ ,  $p = 0.741$ , in 2007;  $H = 19.19$ ,  $p = 0.1388$  in 2016/17; Tables 1 and 2).

### 3.2. $\delta^{15}\text{N}$ as a proxy of trophic structure

$\delta^{15}\text{N}$  values for each species in 2007/08 and 2016/17 are given in Tables 1 and 2, respectively.  $\delta^{15}\text{N}$  was significantly higher in seabirds (7.69 ‰ in Antarctic prions to 15.50 ‰ in wandering albatrosses (*Diomedea exulans*) and notothenioid fish (only in 2016/17; 7.71 ‰ in *Patagonotothen guntheri* to 14.41 ‰ in *N. rossii*) than in the other taxonomic groups (Kruskal-Wallis test,  $H = 59.184$ ,  $p < 0.0001$ ,  $H = 80.284$ ,  $p < 0.0001$  for birds and fish respectively). Moreover, values decreased from myctophid fish (6.69 ‰ in *E. antarctica* to 10.91 ‰ in *G. opisthopterus*), to squid (5.68 ‰ in *S. circumantarctica* to 9.38 ‰ in *Galiteuthis glacialis*) and then zooplankton (1.73 ‰ in *E. superba* to 8.81 ‰ in *Gigantocypris* sp.).

In 2007/08, there were significant differences in  $\delta^{15}\text{N}$  values among seabird species (Kruskal-Wallis test,  $H = 20.979$ ,  $p = 0.0008$ , Table 1). Wandering albatrosses had significantly higher  $\delta^{15}\text{N}$  than black-browed albatrosses (*Thalassarche melanophris*; Dunn's multiple comparisons test,  $p = 0.0043$ ), grey-headed albatrosses (*Thalassarche chrysostoma*; Dunn's multiple comparisons test,  $p = 0.0340$ ) and southern giant petrels (*M. giganteus*; Dunn's multiple comparisons test,  $p = 0.0049$ ). In 2016/17, there were also differences among seabird species (Kruskal-Wallis test,  $H = 21.756$ ,  $p = 0.001$  Table 2) with Antarctic prions having significantly lower  $\delta^{15}\text{N}$  values than brown skuas (Dunn's multiple comparisons test,  $p = 0.0251$ ) and grey-headed albatrosses (Dunn's multiple comparisons test,  $p = 0.0202$ ).

$\delta^{15}\text{N}$  values also differed significantly among species of notothenioid fish (Kruskal-Wallis test,  $H = 25.847$ ,  $p < 0.001$ , Table 2) with *C. gunnari* and *P. guntheri* having significantly lower values than *N. gibberifrons* (Dunn's multiple comparisons test,  $p = 0.0045$ ;  $p = 0.0273$ ) and *D. eleginoides* (Dunn's multiple comparisons test,  $p = 0.0029$ ;  $p = 0.0124$ ). No significant differences in  $\delta^{15}\text{N}$  were detected between species of myctophid fish (Kruskal-Wallis test,  $H = 7.996$ ,  $p = 0.238$ , in 2007/08;  $H = 12.888$ ,  $p = 0.244$ ; Tables 1 and 2) and squid (Wilcoxon rank test,  $W = 11$ ,  $p = 0.393$ , in 2007/08;  $W = 6$ ,  $p = 0.200$  in 2016/17; Tables 1 and 2) in either of the sampling years. Significant differences in  $\delta^{15}\text{N}$  were only observed between two species of zooplankton (Kruskal-Wallis test,  $H = 20.03$ ,  $p = 0.010$ ; Table 2), *E. superba* and *P. boeckii* in 2007/08 (Dunn's multiple comparisons test,  $p = 0.0032$ ).

### 3.3. Mercury concentrations

In both sampling years, seabirds had the highest Hg concentrations among the analysed species. In seabirds, Hg concentrations ranged from 0.12  $\mu\text{g g}^{-1}$  in Antarctic prions to 7.17  $\mu\text{g g}^{-1}$  in brown skuas; the next highest values were in myctophid fish (0.025  $\mu\text{g g}^{-1}$  in *Krefflichthys anderssoni* to 0.352  $\mu\text{g g}^{-1}$  in *G. nicholsi*) and notothenioid fish (0.007  $\mu\text{g g}^{-1}$  in *C. gunnari* to 0.343  $\mu\text{g g}^{-1}$  in *N. rossii*), squid (0.012  $\mu\text{g g}^{-1}$  in *S. circumantarctica* to 0.066  $\mu\text{g g}^{-1}$  in *G. glacialis*) and zooplankton (0.006  $\mu\text{g g}^{-1}$  in *E. superba* to 0.141  $\mu\text{g g}^{-1}$  in *P. boeckii*). As expected, POM had the lowest Hg concentrations (0.0005  $\pm$  0.0002  $\mu\text{g g}^{-1}$ ). Highly significant differences were found in total Hg concentrations between taxonomic groups (including POM), within both sampling years (Kruskal-Wallis test,  $H = 59.75$ ,  $p < 0.0001$ , in 2007/08;  $H = 82.42$ ,  $p < 0.0001$

in 2016/17).

Significant differences in Hg concentrations were observed among seabird species both in 2007/08 (Kruskal-Wallis test,  $H = 22.621$ ,  $p = 0.0003$ ; Table 1) and 2016/17 (Kruskal-Wallis test,  $H = 24.175$ ,  $p = 0.0005$ ; Table 2). In 2007/08, wandering albatrosses showed the highest concentrations, followed by brown skuas, grey-headed albatrosses, black-browed albatrosses, southern giant petrels and northern giant petrels. In 2016/17, brown skuas had the highest concentrations followed by northern giant petrels, southern giant petrels, black-browed albatrosses, grey-headed albatrosses, blue petrels and Antarctic prions.

In notothenioid fish, there were highly significant differences between species (Kruskal-Wallis test,  $H = 27.795$ ,  $p < 0.0001$ ). In particular, *C. gunnari* had significantly lower Hg concentrations than *N. gibberifrons*, *N. rossii* and *D. eleginoides* (Dunn's multiple comparisons test,  $p = 0.0004$ ;  $p = 0.0009$ ,  $p = 0.0028$ ; Table 2). Myctophids showed differences between the species in both sampling years (Kruskal-Wallis test,  $H = 15.317$ ,  $p = 0.018$ , in 2007/08;  $H = 13.52$ ,  $p = 0.019$  in 2016/17; Tables 1 and 2), with *G. nicholsi* having a higher Hg concentration than *P. bolini* (Dunn's multiple comparisons test,  $p = 0.0368$  in 2007/08;  $p = 0.0375$  in 2016/17) and *K. anderssoni* (Dunn's multiple comparisons test,  $p = 0.0325$  in 2007/08;  $p = 0.0381$  in 2016/17). There were no significant differences in Hg concentrations between squid species in either year (Wilcoxon rank test,  $W = 15$ ,  $p = 0.057$ , in 2007/08;  $W = 4$ ,  $p = 0.800$  in 2016/17) nor among zooplankton species in 2007/08 (Kruskal-Wallis test,  $H = 7.4231$ ,  $p = 0.492$ ). However, in 2016/17, there were significant differences between species in 2016/2017 (Kruskal-Wallis test,  $H = 20.445$ ,  $p = 0.0088$ ).

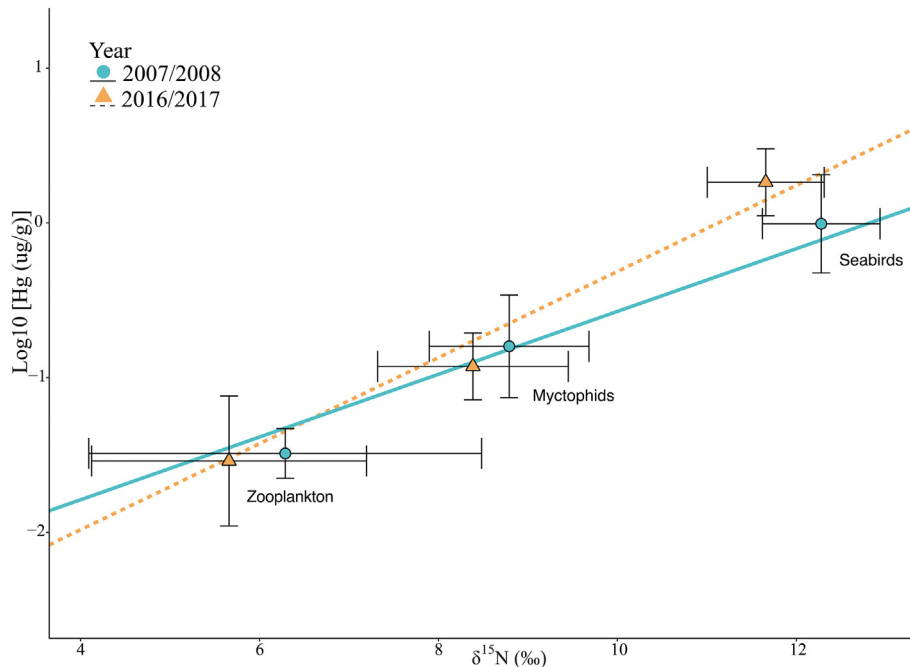
Significant positive linear regressions were found between  $\log_{10}\text{Hg}$  concentrations and  $\delta^{15}\text{N}$  values across all species in both years ( $Y = 0.2112 * X - 2.8088$ ,  $r^2 = 0.7904$ ,  $p < 0.0001$  in 2007/8;  $Y = 0.2451 * X - 3.0363$ ,  $r^2 = 0.8419$ ,  $p < 0.0001$  in 2016/17).

### 3.4. Comparison between years

Five of the eight seabird species were sampled in both study years (brown skuas, grey-headed and black-browed albatrosses, northern and southern giant petrels). With regards to  $\delta^{15}\text{N}$ , significant differences between 2007/08 and 2016/17 values were only found for northern giant petrels ( $t$ -test,  $t_8 = 3.907$ ,  $p = 0.0045$ ), whereas  $\delta^{13}\text{C}$  values differed in most seabirds with the exception of northern giant petrels ( $t$ -test,  $t_9 = 1.693$ ,  $p = 0.1246$ ). Hg concentrations were generally higher in 2016/17 than in 2007/08, but this was significant only for northern giant petrels (Mann Whitney test,  $U = 0$ ,  $p = 0.0079$ ) and southern giant petrels ( $t_8 = 6.322$ ,  $p < 0.005$ ).

In myctophids, five out of the seven species were caught in both sampling years (*E. antarctica*, *G. nicholsi*, *G. braueri*, *K. anderssoni* and *P. bolini*).  $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$  were similar in all species between the two periods. Unlike seabirds, Hg concentrations were lower in 2016/17 than in 2007/08, and significantly so in the case of *E. antarctica* (Mann Whitney test  $U = 45$ ,  $p = 0.002$ ). Six species of zooplankton were caught in both sampling years (*E. triacantha*, *Parandania boeckii*, *Gigantocypris* sp., *E. superba*, *Thysanoessa* sp., *Themisto gaudichaudii*). No significant differences in  $\delta^{15}\text{N}$ ,  $\delta^{13}\text{C}$  or Hg were detected between any zooplankton species between 2007/08 and 2016/17.

There were significant positive correlations between  $\log_{10}\text{Hg}$  concentrations and  $\delta^{15}\text{N}$  values in the species that were sampled in both years ( $Y = 0.2028 * X - 2.6008$ ,  $r^2 = 0.8138$ ,  $p < 0.0001$  in 2007/8;  $Y = 0.2782 * X - 3.0960$ ,  $r^2 = 0.9314$ ,  $p < 0.0001$  in 2016/17) (Fig. 1). The slope of the relationship was significantly higher in 2016/17 (0.2782) than 2007/08 (0.2028) (ANOVA,  $F_{1, 110} = 9.716$ ,  $P = 0.0023$ ).



**Fig. 1.** Mercury concentration on a log 10 scale versus  $\delta^{15}\text{N}$  for zooplankton (whole individuals), myctophid fish (muscle) and seabirds (feathers) that were caught in 2007/08 ( $Y = 0.2028 \cdot X - 2.6008$ ) and in 2016/17 ( $Y = 0.2782 \cdot X - 3.0960$ ).

#### 4. Discussion

A number of studies have used nitrogen stable isotope values ( $\delta^{15}\text{N}$ ) to contextualise the biomagnification of Hg within food webs in different ecosystems (Cabana and Rasmussen, 1994; Lavoie et al., 2013). However, to the best of our knowledge, this study is the first to evaluate Hg bioaccumulation from POM to top predators in a Southern Ocean food web.

##### 4.1. Influence of feeding habitat

The first assumption when evaluating food web structure and links using stable isotope ratios is that all the analysed species share broadly the same environment. This assumption is required to rule out the potential for any spatial variation in baseline  $\delta^{15}\text{N}$  in apparent trophic level within geographically distinct ecosystems (Chouvelon et al., 2012). Ecosystem connectedness can be determined using  $\delta^{13}\text{C}$  as a proxy for habitat (Kelly, 2000), and all taxonomic groups in our study had broadly similar  $\delta^{13}\text{C}$  values (Tables 1 and 2). The greatest differences in  $\delta^{13}\text{C}$  values were observed in seabirds, which is to be expected as feathers are more enriched in  $^{13}\text{C}$  than blood, muscle and internal organs (Cherel et al., 2014; Kelly, 2000), and we assert that the distinct seabird  $\delta^{13}\text{C}$  values are a consequence of the use of different tissues rather than any spatial separation. This assertion is backed up by tracking data during chick-rearing, which shows that all the sampled seabird species forage broadly in the same areas where the myctophids, squid and zooplankton were caught [wandering albatrosses (Jiménez et al., 2015); southern giant petrels and northern giant petrels (Granroth-Wilding and Phillips, 2018a); brown skuas (Carneiro et al., 2014); Antarctic prions and blue petrel (Navarro et al., 2013); black-browed albatrosses and grey-headed albatrosses (Phillips et al., 2004b)]. As expected, there were small differences in  $\delta^{13}\text{C}$  values among some species within taxonomic groups, indicating a limited degree of divergence in foraging habits (e.g. feeding depth), but this has no material effect on our

conclusions about trophic level based on  $\delta^{15}\text{N}$ . Hg concentrations throughout the sampled food web were not influenced by differences in habitat utilization (inferred from the  $\delta^{13}\text{C}$  values), as our analysis reflects a single regional system in the Scotia Sea in the vicinity of South Georgia.

##### 4.2. Trophic structure in relation to mercury concentrations

Our combined field sampling provided two snapshots, 9 years apart, of Hg concentrations in the Scotia Sea food web. Values of Hg were lowest in POM, followed by zooplankton, which as a group exhibited some degree of variation. This is not surprising since the zooplankton species examined range from predominantly herbivorous Antarctic krill (Quetin and Ross, 1991) to predators such as *Themisto gaudichaudii* (Havermans et al., 2019). Squid and myctophid fish link zooplankton to the top predators (seabirds) and, in terms of Hg dynamics, exhibit some degree of variation between individuals and species in Hg concentrations [for more details see (Seco et al., 2020a; 2020b)]. Generally, the highest Hg concentrations were found in predators higher in the food web (nototheniid fish and seabirds). Our first clear result is therefore that total Hg concentration increases with trophic level (reflected by  $\delta^{15}\text{N}$ ). This relationship was expected as a result of Hg biomagnification through food webs, and has previously been reported by other studies in the Antarctic region (Anderson et al., 2009).

Trophic links to the two groups of top predators analysed here, seabirds and nototheniid fish (the latter taxa are sometimes considered to be mesopredators, as they can themselves be predated by larger species), can be considered as two parallel paths in the Scotia Sea food web, because both groups of top predators occupy similar trophic positions (see Tables 1 and 2). However, our results reveal that the seabirds can be differentiated into three distinct groups with characteristic Hg levels: wandering albatrosses and brown skuas had the highest Hg concentrations of all seabird species ( $1.556\text{--}7.173 \mu\text{g g}^{-1}$ ); grey-headed albatrosses, black-browed albatrosses, southern giant petrels and northern giant

petrels, had intermediate Hg levels ( $0.122\text{--}2.95\ \mu\text{g g}^{-1}$ ), and Antarctic prions and blue petrels had the lowest Hg levels ( $0.120\text{--}0.959\ \mu\text{g g}^{-1}$ ). This is a classic effect of diet on Hg levels in seabirds, as it has been showed before (Blévin et al., 2013; Carravieri et al., 2014). Levels of Hg concentration in seabirds feathers are directly influenced by seabird trophic level: seabirds that feed on zooplankton have lower concentrations than fish- and squid-eating species (Bocher et al., 2003; Carravieri et al., 2014b). Wandering albatrosses feed on higher trophic level prey, mainly large fish and squid (Moreno et al., 2016), which our study shows had relatively high Hg concentrations. Brown skuas have a diverse diet, feeding on Antarctic fur seal *Arctocephalus gazella* carrion (including placentae), other seabirds and occasionally fish or squid which they obtain via kleptoparasitism (Phillips et al., 2004a). Carravieri et al. (2014b) showed that brown skua chicks at Kerguelen Islands have higher Hg feather concentrations than the adults, probably because their diet consists mainly of blue petrels, whereas adults during the non-breeding period consume a more diverse diet, including marine prey. High diet diversity probably also explains the high variability in Hg and  $\delta^{15}\text{N}$  in grey-headed albatrosses and black-browed albatrosses, which feed on fish, squid and crustaceans (Prince, 1980a). Northern giant petrels and southern giant petrels are more generalist, feeding both on carrion on land (from Antarctic fur seals and penguins) and Antarctic krill, squid and other seabirds (Hunter, 1983). Additionally, both species present sex differences in feeding habits, with males predominantly scavenging during the early-mid breeding season, whereas females mostly forage at sea (González-Solís et al., 2000; Granroth-Wilding and Phillips, 2018b). The smaller seabirds in our study, Antarctic prions and blue petrel, feed mainly on zooplankton, including Antarctic krill (Prince, 1980b). They are therefore exposed to lower Hg concentrations than seabirds that are piscivorous or teuthophageous, as reported in other breeding areas (Blévin et al., 2013; Carravieri et al., 2014). Thus, the smaller bird species have Hg concentrations in the same range as some fish and squid species, probably also because chicks have had only a short exposure period, which neglects the potential effect of Hg bioaccumulation. Hg in chick feathers reflect their exposure from the time they were born, to the time of the feather synthesis which is in our case a few months.

The use of seabirds feathers can provide an accurate measurement to the biomagnification of Hg along food webs, as this tissues does not show increasing Hg concentrations with age (Thompson et al., 1991; Donaldson et al., 1997; Fevold et al., 2003; Tavares et al., 2013). Hg is transferred to feathers only when they are synthesised and then the feathers are metabolic inert, meaning their Hg concentration will not vary along time.

Differences in Hg concentrations were also found between the notothenioid fish species. Hg concentrations were lower in *C. gunnari* than in all the other species, which is consistent with the reported results for these fish at the Kerguelen Islands (Bustamante et al., 2003; Cipro et al., 2018). These differences only partly reflect trophic level, as  $\delta^{15}\text{N}$  was similar in *C. gunnari* and *P. guntheri*. Nor does the discrepancy appear to mirror what is known about diet, as both species are considered to feed mostly on euphausiids and amphipods (Collins et al., 2008; di Prisco et al., 1991). Furthermore, the influence of body size can be discounted, as *C. gunnari* were on average 67 mm bigger than *P. guntheri*, and larger fish tend to have higher Hg concentrations in species with similar growth rates (Dang and Wang, 2012; Gewurtz et al., 2011). Instead, the low Hg concentrations seem more likely to relate to the highly specialized physiological characteristics of *C. gunnari*, including the absence of haemoglobin (Sidell and O'Brien, 2006), which is associated with

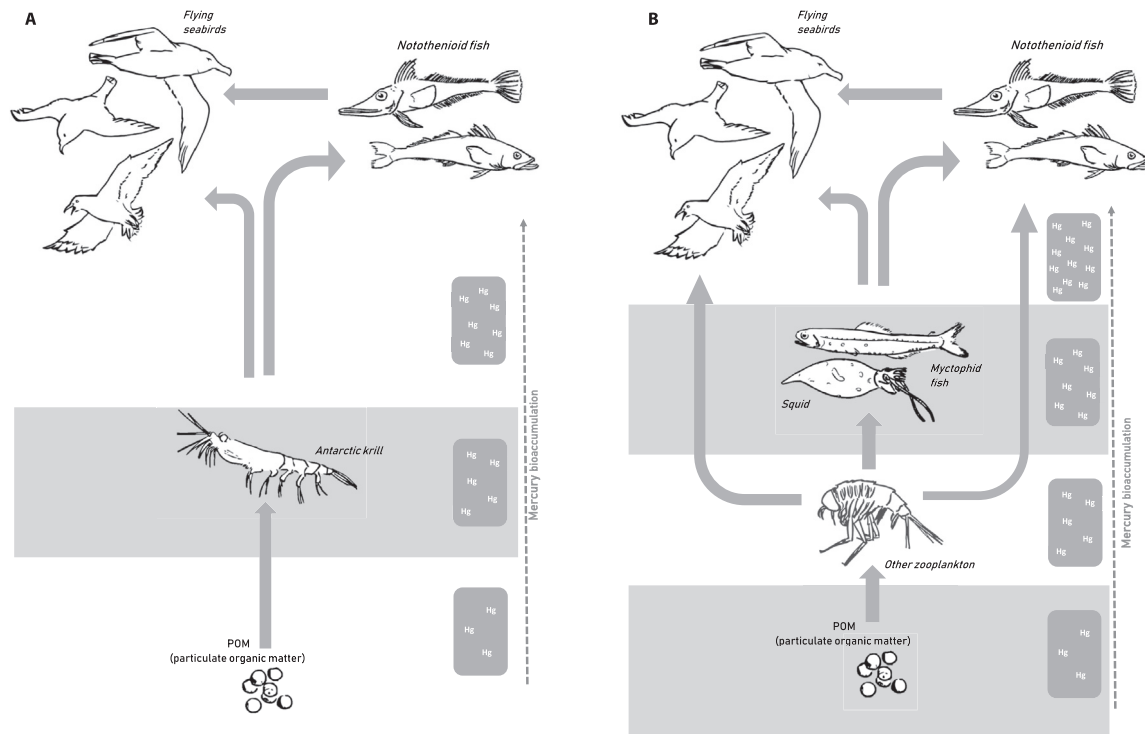
lower metabolic rates (Johnston and Camm, 1987). However, this would need to be confirmed by further studies.

As in seabirds, the notothenioid fish species at higher trophic levels had higher Hg concentrations. *D. eleginoides* feeds mostly on fish, squid and crustaceans (Collins et al., 2007, 2010; Seco et al., 2015). In contrast, *N. gibberifrons* and *N. rossii* feed on diverse prey from algae to amphipods, euphausiids and other fish (Casaux et al., 1990), which would also explain the large variability in their  $\delta^{15}\text{N}$  values. The diet of *C. aceratus* is dominated by crustaceans and fish (Reid et al., 2007). Together, these guild-specific results highlight the important role of diet in Hg bioaccumulation.

The slope of the linear regression between  $\log_{10}(\text{Hg})$  concentration and  $\delta^{15}\text{N}$  values, also known as the trophic magnification slope (TMS), is an indicator of Hg biomagnification potential in a food web (Lavoie et al., 2013; Yoshinaga et al., 1992). Hg TMS can be influenced not only by food web dynamics but also by habitat characteristics (Lavoie et al., 2013). At lower latitude regions, slower growth rates and slower Hg excretion rates, due to colder temperatures, could lead to greater biomagnification of Hg ( $0.21 \pm 0.07$ ; Lavoie et al., 2013) that the at higher latitudes, where higher primary productivity and growth rates (Gross et al., 1988; Pauly, 1998) may lead to lower TMS ( $0.16 \pm 0.08$ ; Lavoie et al., 2013). TMS values for the Scotia Sea ecosystem were 0.267 for 2007/08 and 0.200 for 2016/17, which are both within the range of those previously reported for polar regions (Lavoie et al., 2013).

#### 4.3. Interannual variation in mercury concentration

When comparing species collected in both sampling years, Hg concentrations in the mid trophic-level groups (squid and myctophid) were lower in 2016/17 than in 2007/08, as reported in detail by Seco et al. (2020a,b). In contrast, Hg concentrations were higher in the seabirds sampled in 2016/17 than in 2007/08, which is the opposite pattern to the mid trophic-level species (Seco et al., 2020a). This was unexpected as the Hg body burdens of predators should, in theory, reduce if there is less bioavailable Hg in their prey (Atwell et al., 1998). A plausible explanation is that there was a change in the main trophic pathway between years (Ward et al., 2010). Differences in Hg levels in producers as well as changes in the productivity and food web structure can directly influence Hg concentration in top predators (Ward et al., 2010). The Scotia Sea food web is centred on Antarctic krill (Murphy et al., 2007), the abundance of which can determine the reproductive success and survival of dependent predators (Grünbaum and Veit, 2003; Lynnes et al., 2004; Seyboth et al., 2016). In years with low Antarctic krill abundance, predators switch to alternative food sources, particularly myctophid fish or squid (Murphy et al., 2007; Mills et al., 2020). In our study, Antarctic krill had amongst the lowest Hg concentrations, suggesting that in situations when less krill is available predators, a switch to alternative prey with higher Hg burdens (Fig. 2B) will result in predators being subjected to greater levels of Hg bioaccumulation. Abundance of Antarctic krill based on acoustic surveys to the northwest of South Georgia was relatively low in 2016/17 (BAS, unpublished data) when compared with previous years (Fielding et al., 2014). Our Hg data possibly illustrates the knock-on effect of variable Antarctic krill abundance on food web dynamics, particularly the effect on higher predators that cascade through the food web. This is of particular interest in light of studies that suggest there is a long-term decline in abundance of Antarctic krill in the Scotia Sea (Atkinson et al., 2019; Hill et al., 2019; Rintoul et al., 2018). In years of low Antarctic krill abundance, predators not only have to cope with the stress of reduced prey availability, but with a concomitant increase in Hg exposure.



**Fig. 2.** Illustrations of the South Georgia food web in years with high Antarctic krill abundance (left – A) and in years with low Antarctic krill abundance (right – B). Shaded/clear areas represent different trophic levels, boxes represent mercury concentrations (more Hg – higher concentration).

Our results highlight that changes in food web dynamics, particularly temporal switches between krill-based and non-krill-based trophic pathways, are likely to be important in the transport of Hg between mid-trophic levels and apex predators in the Southern Ocean.

**Author contribution**

José Seco: Writing – original draft; Conceptualization. Collection, identification and preparation of samples for laboratory analysis. Mercury quantification. Sara Aparício: Writing – review & editing, Illustrations and graphical support. Andrew S. Brierley: Writing – review & editing, Supervision, supervisor of the project and financial support. Paco Bustamante: Writing – review & editing, Conceptualization, Supervision, Resources, provided the facilities to stable isotopes analysis of notothenioid fish and help on interpretation of toxicological data. Filipe R. Ceia: Writing – review & editing, stable isotopes protocols analysis for zooplankton, fish, squid and seabirds, interpretation of ecological data. João P. Coelho: Writing – review & editing, Supervision, laboratorial support for mercury analysis, interpretation of toxicological data. Richard A. Philips: Writing – review & editing, provided the seabirds samples and interpretation of sea birds results. Ryan A. Saunders: Writing – review & editing, identification and collection of myctophid fish, provided preserved myctophids samples from 2007/08. Sophie Fielding: Writing – review & editing, identification and collection of zooplankton. Susan Gregory: Resources, provided the notothenioid fish samples. Ricardo Matias: Writing – review & editing, preparation of samples for stable isotopes analysis. Miguel A. Pardal: Writing – review & editing, financial support and conceptualization of the work hypothesis. Eduarda Pereira: Resources,

provided laboratory equipment for mercury analysis. Gabriele Stowasser: Writing – review & editing, identification and collection of zooplankton and squid, provided preserved squid samples from 2007/08. Geraint A. Tarling: Writing – review & editing, Supervision, identification and collection of zooplankton, provided preserved zooplankton samples from 2007/08. José C. Xavier: Writing – review & editing, Conceptualization, Supervision, Resources, Conceptualization of the ecological answer to the toxicology variation, provided the facilities to stable isotopes analysis.

**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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