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Mercury biomagnification in an Antarctic food web of the Antarctic Peninsula☆

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ABSTRACT

Under the climate change context, warming Southern Ocean waters may allow mercury (Hg) to become more bioavailable to the Antarctic marine food web (i.e., ice-stored Hg release and higher methylation rates by microorganisms), whose biomagnification processes are poorly documented. Biomagnification of Hg in the food web of the Antarctic Peninsula, one of the world's fastest-warming regions, was examined using carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) stable isotope ratios for estimating feeding habitat and trophic levels, respectively. The stable isotope signatures and total Hg (T-Hg) concentrations were measured in Antarctic krill *Euphausia superba* and several Antarctic predator species, including seabirds (gentoo penguins *Pygoscelis papua*, chinstrap penguins *Pygoscelis antarcticus*, brown skuas *Stercorarius antarcticus*, kelp gulls *Larus dominicanus*, southern giant petrels *Macronectes giganteus*) and marine mammals (southern elephant seals *Mirounga leonina*). Significant differences in $\delta^{13}C$ values among species were noted with a great overlap between seabird species and *M. leonina.* As expected, significant differences in $\delta^{15}N$ values among species were found due to interspecific variations in diet-related to their trophic position within the marine food web. The lowest Hg concentrations were registered in *E. superba* (0.007 ± 0.008 μg g^{-1}) and the highest values in *M. giganteus* (12.090 ± 14.177 μg g^{-1}). Additionally, a significant positive relationship was found between Hg concentrations and trophic levels (reflected by $\delta^{15}N$ values), biomagnifying nearly 2 times its concentrations at each level. Our results support that trophic interaction is the major pathway for Hg biomagnification in Southern Ocean ecosystems and warn about an increase in the effects of Hg on long–lived (and high trophic level) Antarctic predators under climate change in the future.

1. Introduction

Marine pollution is a growing environmental problem in today's world (Häder et al., 2020). In Antarctica and the Southern Ocean, the effects of anthropogenic activities are visible and threaten the sustainability of this region's delicate ecosystems (Lohmann et al., 2006; Gutt et al., 2020). Mercury (Hg) is a non-essential metal known for its toxicity even at low concentrations, particularly in its methylated form (MeHg), and it has been acknowledged as a global toxicity problem in aquatic ecosystems (Jackson, 1998). Once thought to be pristine, the Antarctic region constitutes a sink for various pollutants such as Hg which condensate in colder regions after evaporating and undergo long–range

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transportation from lower latitudes (Dommergue et al., 2010; Angot et al., 2016). Consequently, the Southern Ocean presents some of the highest MeHg concentrations ever recorded in open waters (Cossa et al., 2011). Even though anthropogenic emissions are negligible (Aronson et al., 2011), the volcanic activity in Antarctic Peninsula significantly contributes to a higher Hg bioavailability in this region (Mão de Ferro et al., 2014). Besides the above-mentioned pathways, the release of Hg stored in sea ice also contributes to its availability for microbial methylation (Cossa, 2013; Gionfriddo et al., 2016). When in the marine environment, Hg can be methylated by microorganisms and then bioconcentrated by phytoplankton and bioaccumulated by organisms along the food chain (Bargagli et al., 1998). MeHg in prey is bioavailable for predators and strongly retained in marine organisms once assimilated, it leads to the stepwise increase of Hg concentrations along the food chain (so-called biomagnification) and to its bioaccumulation with age in organisms (Wiener et al., 2007; Tavares et al., 2013). Previous studies have demonstrated a trend for increasing Hg concentrations from producers < primary consumers < secondary consumers (Dehn et al., 2006; dos Santos et al., 2006; Eagles-Smith et al., 2018; Rodrigues et al., 2019). In the Southern Ocean, previous studies have focused on describing Hg concentrations in specific taxonomic groups (Szefer et al., 1993; Seco et al., 2020), even finding higher Hg concentrations than expected relative to other organisms from more polluted regions, particularly in long-lived predators (Tavares et al., 2013; Goutte et al., 2014b; Cherel et al., 2018).

The use of quantitative Hg analysis coupled with other techniques, such as stable isotope analysis (SIA), can provide a multi-dimensional view of Hg links with ecological traits within marine ecosystems (Lavoie et al., 2013). In order to assess Hg concentrations within a food web, it is essential to assess the trophic levels of their various organisms. Indeed, to our knowledge, few studies are available on Hg biomagnification within Antarctic Peninsula food webs (dos Santos et al., 2006; Cipro et al., 2017), a region that is affected by several threats such as anthropogenic pollution (Tin et al., 2009; Aronson et al., 2011) and climate change impacts (Constable et al., 2014; Gutt et al., 2020; Pertierra et al., 2020). Stable isotope ratios of carbon ($^{13}C/^{12}C$, $\delta^{13}C$) and nitrogen ($^{15}N/^{14}N$, $\delta^{15}N$) are a very useful method to determine feeding habitat and trophic level of organisms, respectively (Cherel & Hobson, 2005; Zimmer et al., 2007). In a marine context, the $\delta^{13}C$ signatures enable the characterization of the organisms' preferential habitat as it varies according to latitude, depth and coast proximity (Cherel & Hobson, 2007; Jaeger et al., 2010). $\delta^{15}N$ signatures can be used as an indicator of consumers' trophic position as ^{15}N values consistently increase from lower to higher trophic levels within a range of 2.5–3.4‰ per trophic level (Minagawa & Wada, 1984). According to the considered tissues and their respective stable isotope turn-overs, stable isotopes can provide short- and long-term environmental signatures of the individual organisms (Ceia et al., 2012) and become a useful tool for reconstructing trophic interactions, elucidating patterns of resource allocation and constructing food webs (Hobson & Welch, 1992; Cherel & Hobson, 2005). Even though several studies have addressed the biomagnification of Hg within food webs resorting to $\delta^{15}N$ values as a proxy of trophic levels, only recently these have been used to determine Hg biomagnification along trophic levels and pathways in Southern Ocean food webs (Anderson et al., 2009; Cipro et al., 2017; Seco et al., 2021).

In this study, we analysed total Hg concentrations, $\delta^{13}C$ and $\delta^{15}N$ values in the tissues of key functional organisms from Livingston Island, South Shetlands Archipelago (Fig. 1), from zooplankton to top predators foraging in marine waters around the west Antarctic Peninsula (Muelbert et al., 2013; Herman et al., 2017). The main objectives of this study, based on samples from Antarctic krill, seabirds and marine mammals were to: 1) characterise the Hg concentrations in the considered organisms and their trophic levels; 2) evaluate the Hg biomagnification levels in the marine food web; and 3) discuss Hg pathways taking place in ecosystems around west Antarctic Peninsula.

2. Materials & methods

2.1. Study area and species

The study area was Livingston Island (62°36′S, 60°30′W), which is the second largest island in the South Shetland Archipelago, in maritime Antarctica (Fig. 1). Regarding climate, average air temperatures are slightly above 0 °C during summer months, dropping below −25 °C during winter. For that reason, ice-free areas in the island host colonies of various seabird and seal species.

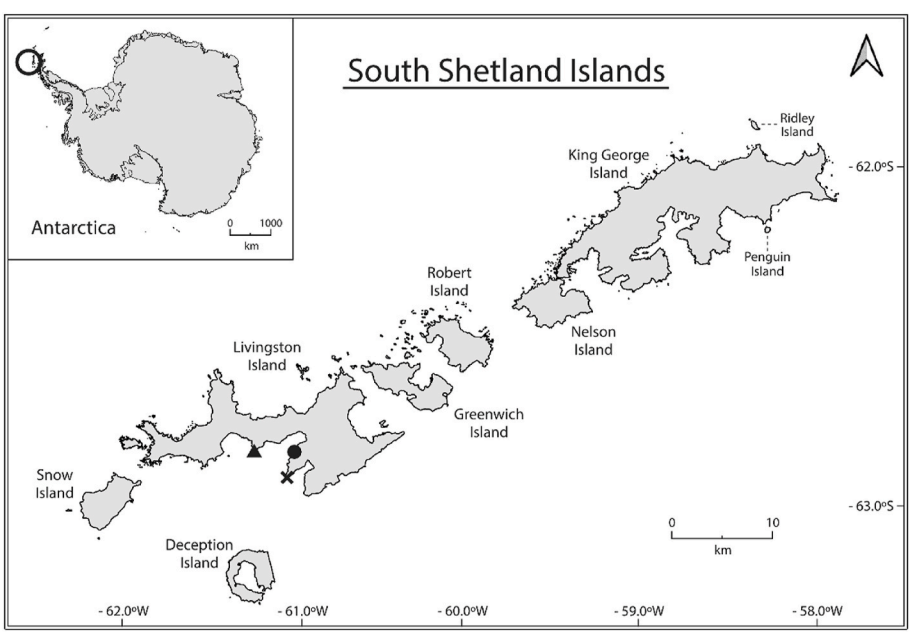

Fig. 1. Location of the Livingston Island in the South Shetland Archipelago, in the northeast Antarctic Peninsula. The sample collection took place in Hannah Point (represented by a triangle), Miers Bluff Point (represented by a cross) and close to the Bulgarian Antarctic base St. Kliment Ohridski research station (represented by a circle).

The Antarctic krill (*Euphausia superba*) was selected as it is keystone species in the Southern Ocean food webs, linking primary producers and predators (Ainley & DeMaster, 1990; Everson, 2000). For this study, several predator species inhabiting the study region that rely directly on Antarctic krill as an important food source were selected, namely gentoo penguin (*Pygoscelis papua*), chinstrap penguin (*Pygoscelis antarcticus*), brown skua (*Stercorarius antarcticus*), kelp gull (*Larus dominicanus*), southern giant petrel (*Macronectes giganteus*) and southern elephant seal (*Mirounga leonina*). Regarding diets, *P. antarcticus* tend to specialise in *E. superba*, whereas *P. papua* has a mixed diet of both krill and fish (Herman et al., 2017). Moreover, *M. leonina* presents a fish and squid-dominated diet (Bradshaw et al., 2003), while the other mentioned seabird species – *L. dominicanus*, *M. giganteus* and *S. antarcticus* – are highly opportunistic, feeding on zooplankton, molluscs, fish and even carrion of seabirds and marine mammals (Hunter, 1983; Reinhardt et al., 2000; Silva et al., 2001).

2.2. Collection of biological samples

Samples were collected in the South Bay of Livingston Island, specifically at Hannah Point, Miers Bluff Point and nearshore close to the Bulgarian Antarctic base St. Kliment Ohridski research station, during January 2011 (austral summer). Specimens of *E. superba* (n = 62, pooled samples) were collected from the stomachs of dissected *P. papua* and *P. antarcticus*, and from black rockcod *Notothenia coriiceps* and marbled rockcod *N. rossii*, two common fish species in this region (Uzunova et al., 2020). Rockcod fish specimens were collected by pole fishing at the South Bay, near the research station. Back feathers from *P. papua* (n = 30) and *P. antarcticus* (n = 30) were collected from randomly selected adult specimens returning to the colonies located in Hannah Point, Miers Bluff Point and near the research station. Back feathers from *S. antarcticus* (n = 5), *L. dominicanus* (n = 13) and *M. giganteus* (n = 15), and hair samples from *M. leonina* (n = 15) were collected from randomly selected adult specimens aggregated in Hannan Point. All samples were preserved in individual bags at room temperature, with the exception of *E. superba*, which were preserved at −20 °C until further analysis.

2.3. Stable isotope and mercury analyses

The SIA protocol was performed in the MAREFOZ laboratory in Figueira da Foz, Portugal. Samples of *E. superba* were ground into a fine powder and its lipidic content was extracted using cyclohexane, due to its depletion in ^{13}C relative to other tissue components. Back feather and hair samples were cleaned of surface contaminants using a 2:1 solution of chloroform-methanol and air-dried in an oven at 60 °C for 48 h. Thereafter, feather and hair samples were cut with a stainless-steel scissor into small fragments and discarded the calamus in feathers. After homogenising all samples, approximately 0.35 mg of each sample was analysed for $\delta^{13}C$ and $\delta^{15}N$ values using a Continuous Flow Isotope Mass Spectrometer (CF-IRMS) coupled with an elemental analyser. Ratios were calculated using the formula, $\delta X = [(R_{Sample}/R_{Standard}) - 1] \times 1000$, considering the standardised values of Vienna-PeeDee Belemnite (VPD-B) and atmospheric N_2 for $\delta^{13}C$ and $\delta^{15}N$ values, respectively (Hobson & Welch, 1992). The stable analysis on fish were not carried out due to insufficient material collected in the field.

The total mercury (T-Hg) analysis was performed in the CESAM laboratory in the University of Aveiro, Portugal. For the T-Hg analysis were used a small number of specimens (n = 15) of *E. superba*, *P. papua* and *P. antarcticus* than for SIA (see *n* size in Table 1). The total Hg concentrations were determined to the nearest 0.01 ng by thermal decomposition atomic absorption spectrometry with gold amalgamation, using an Advanced Mercury Analyser AMA LECO 254, following Costley et al. (2000). Analyses were done several times until the coefficient of variation was lower than 10%. Accuracy was checked using certified reference materials (CRM) at the beginning of each sample. The CRM used and respective recoveries were dogfish muscle DORM–2 (98.1

Table 1
Total mercury (Hg) concentrations, nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) stable isotope signatures, and C/N ratios registered of Antarctic species collected in the Livingston Island, South Shetland Archipelago in the west Antarctic Peninsula, during the austral summer of 2011.

Species	Tissue	*n*	[Hg] ($\mu g \cdot g^{-1}$ dw)				$\delta^{15}N$ (‰)				$\delta^{13}C$ (‰)				C/N			
			Mean	Median	SD	min, max	Mean	Median	SD	min, max	Mean	Median	SD	min, max	Mean	Median	SD	min, max
Euphausia superba	whole	15	0.007	0.004	0.008	0.003, 0.015	5.05	4.95	0.56	4.09, 5.78	−24.81	−24.64	0.89	−26.55, −23.51	3.22	3.23	0.07	3.04, 3.31
Pygoscelis papua	feathers	15	0.220	0.190	0.092	0.112, 0.380	9.15	9.25	0.64	7.60, 9.89	−22.97	−22.64	0.74	−24.22, −21.54	3.21	3.17	0.20	3.09, 3.93
Pygoscelis antarcticus	feathers	15	0.665	0.517	0.464	0.239, 1.574	8.66	8.98	0.96	7.23, 9.93	−23.15	−23.22	0.62	−24.19, −22.03	3.18	3.18	0.04	3.10, 3.25
Stercorarius antarctius	feathers	5	1.104	0.625	0.785	0.516, 2.288	12.88	11.67	3.36	9.78, 17.91	−17.89	−17.22	1.77	−19.93, −15.73	3.14	0.32	0.04	3.10, 3.23
Mirounga leonine	hairs	15	1.654	0.734	1.390	0.867, 2.998	12.20	12.23	0.86	10.72, 13.86	−21.81	−21.79	0.35	−22.61, −21.31	3.18	0.32	0.04	3.13, 3.26
Larus dominicanus	feathers	13	10.755	10.056	6.750	2.646, 22.486	13.49	13.50	1.28	11.50, 15.42	−21.68	−22.04	1.09	−22.88, −19.34	3.18	0.31	0.04	3.12, 3.23
Macronectes giganteus	feathers	15	12.090	6.497	14.177	1.912, 54.680	14.29	14.18	1.69	11.60, 18.30	−20.30	−21.68	2.23	−22.64, −16.37	3.18	0.32	0.04	3.13, 3.26

± 1.2%), mussel tissue NIST2976 (100.1 ± 12.6%) and lobster hepatopancreas TORT–2 (100.3 ± 9.8%). All Hg data is expressed as a function of dry weight (μg•g^{-1} dw).

Despite being metabolic inert, feathers and hairs are the main excretion route for Hg bioaccumulated in internal tissues since the previous moult (Dietz et al., 2009; Peterson et al., 2016; Albert et al., 2019). Nevertheless, even though both tissues may range within similar values, underlying differences in both tissues' physiology and turnover rates should be taken into account when comparing stable isotope signatures and Hg levels, particularly between hair and feather samples whose relationship remains a gap of knowledge (dos Santos et al., 2006; Dietz et al., 2009). Their Hg concentrations, δ^{13}C and δ^{15}N values do not vary much over time and are generally in line with the diet and foraging habitat of seabirds, providing a perspective on the species Hg annual exposure (Ling, 2013; Carravieri et al., 2014; Albert et al., 2019). Regarding both penguin species (*P. papua* and *P. antarcticus*) and *M. leonina*, stable isotopes in feathers and hairs reflect the environment during the moulting period between mid-March and mid-April each year. During this period, both follicle types actively synthesise feathers and hairs and excrete Hg bioaccumulated, before becoming inactive until the next moulting period in the following year (Carravieri et al., 2013; Ling, 2013). Moreover, the remaining seabird species – *S. antarcticus*, *L. dominicanus* and *M. giganteus* – moult far away from breeding grounds in an asynchronous pattern and during a period extending to several months (Kopp et al., 2011; Carravieri et al., 2014; Krietsch et al., 2017). Hence, we acknowledge that some Hg contamination in the latter species might originate from outside areas.

2.4. Statistical analysis

Linear relationship between logarithm–transformed Hg concentrations ($\log_{10}$[Hg]) and ^{15}N values was used to evaluate Hg biomagnification along trophic levels according to Nfon et al. (2009). The trophic magnification factor (TMF) was calculated using the formula:

$$TMF = 10^{b}$$

where *b* is the slope of the linear relationship between the $\log_{10}$[Hg] and δ^{15}N values, also known as trophic magnification slope (TMS). TMF represents the increase of Hg concentrations per trophic level (TL). The TL of a consumer was estimated by:

$$TL_{Consumer} = (\delta^{15}N_{Consumer} - \delta^{15}N_{Baseline})/\Delta^{15}N + \lambda$$

where $TL_{Consumer}$ is the trophic level of a given consumer and $\delta^{15}N_{Consumer}$ its δ^{15}N values (Lavoie et al., 2013). The $\delta^{15}N_{Baseline}$ is the δ^{15}N values for the baseline organism – in this study, *E. superba* – and its trophic level (λ, TL = 2 for primary consumers). Δ^{15}N is the trophic discrimination factor used for ^{15}N equal to 3.4‰, which is the most frequently used for marine studies (Borgå et al., 2012).

The data normality was assessed using Shapiro–Wilks's test and, due to the non-nominal distribution of the data, statistical differences among species' Hg concentrations, δ^{13}C and δ^{15}N values were assessed by Kruskal–Wallis test and Dunn's post hoc test for multiple species' comparison. *P*–values were considered statistically significant when $p < 0.05$. All the calculations were performed using GraphPad (La Jolla, California, USA) and R statistical software (R Core Team, 2020).

3. Results

3.1. Carbon and nitrogen stable isotopes across the marine food web of Antarctic Peninsula

Species differed significantly (Kruskal–Wallis test; H = 129.4, $p < 0.001$) regarding δ^{13}C values, with *E. superba* presenting the lowest mean values (−25.30 ± 1.29‰, n = 62) and significantly differing from the rest of the species. *P. papua* values of δ^{13}C (−22.78 ± 0.77‰, n = 30) only significantly differed from *E. superba* values ($p < 0.001$). Also, δ^{13}C values of *P. antarcticus* (−23.36 ± 0.67‰, n = 29) were significantly lower than the values registered in *S. antarcticus* (−17.89 ± 1.77‰, n = 5; p = 0.011), *L. dominicanus* (−21.68 ± 1.09‰, n = 13; p = 0.050), *M. giganteus* (−20.30 ± 2.23‰, n = 15; p = 0.001) and *M. leonina* (−21.81 ± 0.35‰, n = 15; p = 0.006), which not differed between them. Only the δ^{13}C values of *E. superba* ($p < 0.001$) and *P. antarcticus* were significantly lower than *S. antarcticus*.

Significant differences were also found among species in δ^{15}N values (Kruskal–Wallis test; H = 149.7, $p < 0.001$). As expected, the lowest δ^{15}N values found in *E. superba* (5.20 ± 0.72‰, n = 62) differed significantly from the rest of the species. An intermediate level of δ^{15}N values was occupied by *P. papua* (8.69 ± 0.82‰, n = 30) and *P. antarcticus* (8.66 ± 0.88‰, n = 29), both differing from the higher values registered in *L. dominicanus* (p = 0.013 and p = 0.009, respectively; 13.49 ± 1.28‰) and *M. giganteus* (p = 0.002 and p = 0.001, respectively; 14.29 ± 1.68‰). Both penguin species' δ^{15}N values did not differ from *M. leonina*. Only, *E. superba* δ^{15}N values differed from *S. antarcticus* ($p < 0.001$).

3.2. Mercury levels and its biomagnification levels in the food web of Antarctic Peninsula

Significant differences in Hg concentrations were found among species (Kruskal–Wallis test; H = 83.3, $p < 0.001$), with overall values ranging from 0.003 μg g^{-1} dw in *E. superba* to 54.680 μg g^{-1} dw in *M. giganteus* (Table 1). *E. superba* Hg concentrations were the lowest, followed by *P. papua*, *P. antarcticus*, *S. antarcticus* and *M. leonina*. The highest mean values were recorded in *L. dominicanus* and *M. giganteus* (Table 1). The Hg concentrations in *E. superba* were significantly different from the ones found in *P. antarcticus* (p = 0.037), *L. dominicanus* ($p < 0.001$), *M. giganteus* ($p < 0.001$ and p = 0.002, respectively; Table 1).

A significant positive linear relationship ($r^2 = 0.799$, H = 362.1, $p < 0.001$) was found between the $\log_{10}$[Hg] and the δ^{15}N values found in Antarctic species (Fig. 2). The linear relationship can be translated by the equation $\log_{10}[Hg] = 0.2902(\delta^{15}N) - 3.285$, where 0.2902 represents the TMS value (Fig. 2). Regarding TL and considering *E. superba* as our baseline value (TL = 2.00), TL values of secondary consumers were calculated *P. antarcticus* (3.06), *P. papua* (3.20), *M. leonina* (4.16), *S. antarcticus* (4.30), *L. dominicanus* (4.48) and *M. giganteus* (4.72). Between trophic levels, it was calculated a TMF equal to 1.95 ± 1.04 based on δ^{15}N values for a finer analysis.

4. Discussion

4.1. Habitat evaluation

This research provided a snapshot of habitat, trophic levels and Hg concentrations in an Antarctic Peninsula marine ecosystem by focusing on several species that co-habitat during the austral summer. The lowest δ^{13}C values were found in *E. superba*, similar to those reported previously (Brault et al., 2018) and were significantly lower than from top predators (seabirds and southern elephant seals). This may be due to different foraging ecology between *E. superba* and the higher predator, the small enrichments occurring along the food web can be negligible (on average + 1‰ per trophic level; Post (2002)). Stable isotope values can greatly vary among taxonomic groups and within organisms (Cherel et al., 2014; Dimitrijević et al., 2018), thus a careful interpretation of the results must be considered.

No significant differences in δ^{13}C values were found among top predators, with the exception between *P. antarcticus* vs. *S. antarcticus*. Such specific differences might be explained not only by the different isotope turnover rates in tissues (i.e., rate at which tissues replace their isotopic signature by stable isotopes derived from the diet; Hobson and

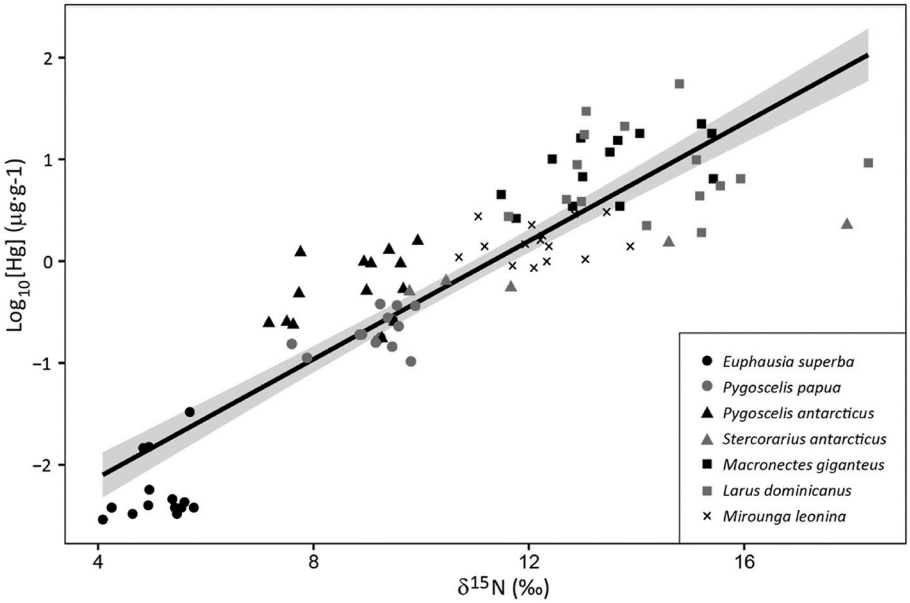

Fig. 2. Linear relationship between logarithm-transformed total mercury (Hg) concentrations (in µg•g^{-1} dw) and nitrogen stable isotope ratios (δ^{15}N, in ‰) recorded in the studied Antarctic species: *Euphausia superba* (n = 15), *Pygoscelis papua* (n = 15), *P. antarcticus* (n = 15), *Stercorarius antarcticus* (n = 5), *Larus dominicanus* (n = 13), *Macronectes giganteus* (n = 15) and *Mirounga leonina* (n = 15). The relationship is given by the solid line (y = 0.2902× − 3.285) and the grey area represents the 95% confidence intervals.

Clark (1992)) but also by the very different species' trophic ecology: *P. antarcticus* feeds mostly on euphausiid, amphipod and myctophid fish species (Rombolá et al., 2010) whereas *S. antarcticus* is highly opportunistic, feeding on carrion and even preying seabirds and pinnipeds (usually chicks, pups and sick/injured adults (Hunter, 1983; Reinhardt et al., 2000)). Stable isotope signatures in feathers are in line with the ecology at their time of synthesis, so variations in δ^{13}C can also be due to different predators' foraging areas as well as different annual migratory patterns to sub-Antarctic and subtropic regions, such as *M. giganteus* and *S. antarcticus*, respectively. However, since our study focuses on understanding Hg dynamics along the food web, δ^{13}C values were only considered as a general indication of species inhabiting the region at the time of the synthesis of the tissue sample collected.

4.2. Relationships between mercury and trophic levels in the Antarctic Peninsula marine food web

The results show a positive relationship between Hg concentrations and δ^{15}N values, as reported by previous studies in the Southern Ocean (Anderson et al., 2009; Chiang et al., 2021; Seco et al., 2021). Based on our TL calculations, a total of three distinct trophic levels were defined throughout the species analysed in this study. As noted in our results, *E. superba* showed the lowest δ^{15}N values due to its primary consumer ecological role, mostly feeding on phytoplankton in open waters (Schmidt et al., 2006). For that reason, *E. superba* occupies the 2nd trophic level among species studied. Both penguin species *P. papua* and *P. antarcticus* occupy the 3rd trophic level, presenting higher δ^{15}N values and Hg concentrations than *E. superba*. These results were expected due to their known trophic ecology with a diet composed of zooplankton, particularly *E. superba*, fish and squid (Xavier et al., 2018; Bertolin & Casaux, 2019). Amongst high predators, *P. papua* and *P. antarcticus* showed the lowest δ^{15}N values and Hg concentrations thus occupying a lower level relative to the species – *M. leonina*, *S. antarcticus*, *L. dominicanus* and *M. giganteus* –, which occupied the 4th and highest trophic level within our study (see TL values in results). Even though *M. leonina* and both penguin species present similar diets, the higher TL value reflected by higher δ^{15}N values in *M. leonina* can be possibly explained by the different tissue nature and by a higher percentage of squid prey in its diet (Bradshaw et al., 2003). These results were expected as previous studies have reported some level of variation in Hg concentrations in seabird species influenced by diet and overall trophy ecology (Anderson et al., 2009; Carravieri et al., 2014). For instance, both penguin species *P. papua* and *P. antarcticus* stay all year round in Antarctic waters and are specialist species feeding mainly on krill whereas the other seabird species – *L. dominicanus*, *M. giganteus* and *S. antarcticus* – migrate and exhibit an opportunistic and scavenging foraging behaviour (Hunter, 1983; Reinhardt et al., 2000; Silva et al., 2001). *P. papua* and *P. antarcticus* present a synchronous moulting pattern after the breeding season, whereas the remaining species renew their feathers throughout a longer period (Carravieri et al., 2013; Ling, 2013). *M. leonina* showed similar δ^{15}N values and Hg concentrations relative to *L. dominicanus* and *M. giganteus*, despite the different nature of the tissues analysed and trophic ecology. In summary, our results indicated a significant increase in Hg concentrations of studied species according with their trophic position, thus indicating biomagnification in the pelagic Antarctic food web of the Antarctic Peninsula.

4.3. Mercury pathways around Antarctic Peninsula and elsewhere

Some of the highest Hg concentrations worldwide have been recorded in marine Antarctic biota, particularly in seabirds from the Southern Ocean (dos Santos et al., 2006; Bargagli, 2008), such as the wandering albatross *Diomedea exulans* (from 3.8 up to 34.6 µg g^{-1} in Cherel et al. (2018)). In our study, the higher concentrations of Hg were reported in *M. giganteus* (from 1.9 up to 54.7 µg g^{-1} in Table 1). The biomagnification of Hg has been addressed before by Bargagli et al. (1998), in the Ross Sea, reporting Hg to increase progressively and consistently in several species from different trophic levels. However, to our knowledge, only Cipro et al. (2017) used stable isotopes to assess the influence of food web structure and diet on Hg concentrations in organisms of the Antarctic Peninsula. Our study reports a significant positive linear relationship between δ^{15}N values and Hg concentrations (TMS = 0.290, TMF = 1.950), similar to those registered by Seco et al. (2021) in the Scotia Sea during the austral summers of 2007 (TMS = 0.203, TMF = 1.596) and 2016 (TMS = 0.278, TMF = 1.897). The differences in TMS values can be explained by different Hg bioavailability governed by the physiology of the organisms (Sontag et al., 2019), as well as site-specific variables, such as the proximity to the coast and ice shelves (Bargagli, 2016; Mastromonaco et al., 2017; Seco et al., 2019). Moreover, the northwest Antarctic Peninsula is a region with high volcanic activity (i.e., in Deception and Penguin islands; see Fig. 1), known natural emission sources of heavy metals (Dick, 1991). Our study supports that diet is a major pathway for Hg bioaccumulation in food webs but more studies are needed to fully understand how the taxonomic,

geographic and ecological differences might influence processes driving Hg in the marine ecosystems of the Antarctic Peninsula.

As the Antarctic Peninsula is one of the fastest-warming regions in the Antarctic region (Amesbury et al., 2017; Rintoul et al., 2018), Hg concentrations in marine biota are expected to increase in next years under the current climate change context due to the enhanced Hg methylation and its bioavailability in low–oxygen subsurface waters, which are likely to increase as global sea temperatures rise (Cossa et al., 2011; Blum et al., 2013). Increased Hg exposure is likely to negatively impact marine biota at behaviour and physiological levels, potentially affecting the reproduction and survival of more affected populations (Evers et al., 2008; Tartu et al., 2013; Goutte et al., 2014a; Goutte et al., 2014b). For that reason, Hg impacts on the structure and functional dynamics of the Antarctic food webs are likely to increase and worth of being monitored, particularly in Antarctic top predators already presenting high Hg concentrations relative to other regions (Gutt et al., 2020; Mills et al., 2020).

5. Conclusion

Our study suggests that Hg biomagnification in the coastal environments of the South Shetland Islands, located in the northeast Antarctic Peninsula, has revealed high Hg concentrations in top predators. Our results support that higher Hg concentrations in predators of higher trophic levels suggest biomagnification of Hg in Southern Ocean ecosystems close to the Antarctic continent. Under a climate change context, Hg concentrations are predicted to increase the negative effects in Antarctic organisms, particularly long-lived predators. Even though Hg biomagnification seems to mainly occur through trophic interactions, further research will need to focus on three main issues: a) Assess on-shore/offshore food webs Hg study in the Antarctic Peninsula region to provide a finer resolution for Hg bioaccumulation studies in this highly climate change impacted region, based on a higher number of study species and sampling size; b) Compare Hg pathways in Antarctic food webs, particularly between coastal Antarctic regions (e.g., Antarctic Peninsula, Ross Sea, Prydz Bay) with oceanic northern regions (e.g., South Georgia, Kerguelen, Macquarie) based on their ecological links and c) Compare Hg bioaccumulation with other stressors (e.g., POP's, macro- and microplastic pollution) and their implications ecologically, Such findings will be relevant for the development of policies under the Minimata Convention and the Antarctic Treaty in the near future, in order to mitigate the exposer and effects of this contaminant.

Author statement

Ricardo Matias: Conceptualization, Writing – original draft and Formal analysis; **Hugo Guímaro:** Conceptualization, Writing – original draft; **Paco Bustamante:** Writing – review & editing; José Seco: Methodology, Writing – original draft; **Nesho Chipev:** Methodology; **Joana Fragão:** Writing – original draft; **Sílvia Tavares:** Methodology; **Filipe Ceia:** Formal analysis, Writing – review & editing; **Maria Pereira:** Resources, Funding acquisition; **Andrés Barbosa:** Writing – review & editing; **José Xavier:** Conceptualization, Methodology; Writing – review & editing, Supervision, Funding acquisition.

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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