Mercury levels in Southern Ocean squid: Variability over the last decade

José Seco [a, b, *], José C. Xavier [c, d], Andrew S. Brierley [b], Paco Bustamante [e], João P. Coelho [f], Susan Gregory [c, g], Sophie Fielding [c], Miguel A. Pardal [h], Bárbara Pereira [a], Gabriele Stowasser [c], Geraint A. Tarling [c], Eduarda Pereira [a]

[a] Department of Chemistry and CESAM/REQUIMTE, University of Aveiro, 3810-193, Aveiro, Portugal
[b] Pelagic Ecology Research Group, Scottish Oceans Institute, University of St Andrews, St Andrews, KY16 8LB, Scotland, UK
[c] British Antarctic Survey, NERC, High Cross, Madingley Road, Cambridge, CB3 0ET, UK
[d] MARE—Marine and Environmental Sciences Centre, Department of Life Sciences, University of Coimbra, Calçada Martim de Freitas, 3000-456, Coimbra, Portugal
[e] Littoral Environnement et Sociétés (LIENSs), UMR 7266 CNRS - La Rochelle Université, 2 Rue Olympe de Gouges, 17000, La Rochelle, France
[f] Department of Biology and CESAM, University of Aveiro, 3810-193, Aveiro, Portugal
[g] Government of South Georgia & the South Sandwich Islands, Stanley, Falkland Islands
[h] CFE - Centre for Functional Ecology, Department of Life Sciences, University of Coimbra, Calçada Martim de Freitas, 3000-456, Coimbra, Portugal

HIGHLIGHTS

- Hg decreases over the last decade, in most of the analysed squid species.
- There is an even partitioning of total Hg among squid tissues.
- Organic Hg was higher in muscle, reflecting the role of diet in bioaccumulation.

ARTICLE INFO

ABSTRACT

The concentrations of total and proportions of organic mercury were measured in tissues of 355 individuals of 8 species of Southern Ocean squid (*Alluroteuthis antarcticus, Bathyteuthis abyssicola, Filippovia knipovitchi, Galiteuthis glacialis, Gonatus antarcticus, Kondakovia longimana, Psychroteuthis glacialis* and *Slosarczykovia circumantarctica*). Squid were caught around South Georgia (Scotia Sea) during 5 cruises, between the austral summers of 2006/07 to 2016/17 to evaluate temporal changes in bioaccumulation and tissue partitioning. Total mercury concentrations varied between $4 \, \text{ng} \, \text{g}^{-1}$ and $804 \, \text{ng} \, \text{g}^{-1}$ among all tissues. Net accumulation of mercury in muscle with size was observed in *A. antarcticus, B. abyssicola* and *P. glacialis*, but no relationship was found for *S. circumantarctica* and lower concentrations were observed in larger individuals of *G. glacialis*. Muscle tissues had the highest mercury concentrations in the majority of species, except for *F. knipovitchi* for which the digestive gland contained highest concentrations. In terms of the percentage of organic mercury in the tissues, muscle always contained the highest values (67%–97%), followed by the digestive gland (22%–38%). Lowest organic mercury percentages were found consistently in the gills (9%–19%), suggesting only low levels of incorporation through the dissolved pathway and/or a limited redistribution of dietary organic mercury towards this tissue. Overall, results are indicative of a decreasing trend of mercury concentrations in the majority of analysed species over the last decade. As cephalopods are an important Southern Ocean trophic link between primary consumers and top predators, these changes suggest decreasing mercury levels in lower trophic levels and an alleviation of the mercury burden on higher predators that consume squid.

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* Corresponding author. Department of Chemistry and CESAM/REQUIMTE, University of Aveiro, 3810-193, Aveiro, Portugal.
E-mail address: jseco@ua.pt (J. Seco).

1. Introduction

Evidence suggest that coleoid cephalopods, such as squid and octopods, are one of the groups that are benefitting from environmental change (Halpern et al., 2008), with some cephalopod populations being on the rise at a variety of locations worldwide (Arkhipkin et al., 2015; Doubleday et al., 2016). Within the Southern Ocean, cephalopods are important links between primary consumers and top predators (Clarke, 1996; Collins and Rodhouse, 2006; Seco et al., 2015). They prey mainly on crustaceans (Kear, 1992; Xavier et al., 2018) and are eaten by a wide range of predators including fish, penguins, albatrosses, seals and whales (Mikhalev et al., 1981; Split, 1995; Xavier and Cherel, 2009). In the Southern Ocean, the waters around South Georgia host a variety of squid species. *Galiteuthis glacialis, Gonatus antarcticus, Filippovia knipovitchi, Kondakovia longimana* and *Psychroteuthis glacialis* are oceanic species and some of the most important prey for several predators (such as seabirds, seals and whales (Xavier and Cherel, 2009)), both by number and by mass (Xavier et al., 2018). *Alluroteuthis antarcticus* and *Slosarczykovia circumantarctica* are taken by a wide range of predators although not in high numbers (Collins and Rodhouse, 2006; Xavier and Cherel, 2009). *Bathyteuthis abyssicola* is a deep-sea squid which is rarely found in the diet of predators. Despite their important role in the Antarctic ecosystem, there is still a lack of knowledge about their ecology (Clarke, 1983; Collins and Rodhouse, 2006; Xavier et al., 2018). Moreover, very few studies have focused on the ecotoxicological aspects of Southern Ocean cephalopods (Anderson et al., 2009; Bustamante et al., 1998; Cipro et al., 2018). Their focal position within Southern Ocean food webs means that cephalopods are likely to be vectors of contaminants and could be valuable bioindicators of ecosystem contamination.

Mercury is one of the contaminants that has been acknowledged as a global toxicity problem (Selin, 2009; UNEP, 2013). Due to its high affinity to proteins (Bloom, 1992), mercury is highly bioaccumulative, becoming toxic for marine organisms higher up the food web (Ackerman et al., 2014; Coelho et al., 2010; Dehn et al., 2006). Furthermore, it biomagnifies along food webs, putting long-lived top predators particularly at risk (e.g. (Goutte et al., 2014; Tartu et al., 2014; Tavares et al., 2013). Indeed, there is already evidence that major cephalopod predators have high levels of mercury in their tissues (e.g. Bustamante et al., 2003; Fontaine et al., 2014; Tavares et al., 2013). In terms of bioavailability within food webs, organic mercury (due to its high affinity to Sulphur-based protein groups) is a particularly toxic form of this element which demands further attention.

In a warming world, in which Antarctica is one of the most rapidly changing and vulnerable areas (IPCC et al., 2013; Rintoul et al., 2018; Turner et al., 2014), organic mercury may become more bioavailable due to the combined influence of increased temperature and depletion of oxygen, which favour methylation of the element by microorganisms (Cossa, 2013). It is therefore important to evaluate the impact that these changes have on the bioavailability of mercury in the Southern Ocean, along with any potential temporal trends in bioaccumulation. Fast growing, short-lived (i.e. generally 1−2 year life cycles) organisms such as squid (Arkhipkin, 2004; Boyle and Rodhouse, 2005) are likely to be responsive bioindicators of contaminant variability over time and space.

Cephalopods bioaccumulate mercury from two main sources: seawater and food. Some mercury uptake can occur through the gills during respiration, from seawater, but mercury in prey is considered to be the main intake pathway (Lacoue-Labarthe et al., 2009). To assess the relative importance of the two pathways in the bioaccumulation of mercury, we analysed mercury in three different tissues: 1) muscle, which represents most of the body weight of the animal and is expected to accumulate high levels of organic mercury (Bustamante et al., 2006); 2) gills, responsible for respiration and subjected to a constant water flow, so likely to be a pathway of incorporation of dissolved Hg from seawater; and 3) digestive gland, which is most affected by the dietary pathway (Bustamante et al., 2006; Penicaud et al., 2017; Pierce et al., 2008) and plays a major role in both the metabolism and detoxification of contaminants such as mercury (Bustamante et al., 2006).

In this study, we evaluate the concentrations of total and organic mercury in 8 different squid species (*A. antarcticus, B. abyssicola, F. knipovitchi, G. glacialis, G. antarcticus, K. longimana, P. glacialis, S. circumantarctica*) from South Georgia between the austral summers of 2006/07 and 2016/17. These species were selected due to their different ecological roles in the Southern Ocean ecosystem (Xavier et al., 2018). The specific objectives of this study are: 1) to evaluate the accumulation pattern along with size (a proxy of age) of Antarctic squid, 2) to understand the partitioning of total and organic mercury in different tissues (muscles, gills and digestive gland) and 3) to assess variability and trends in total and organic mercury concentrations of these species over a 10 year period (2006/07 to 2016/17).

2. Material and methods

2.1. Sampling

South Georgia is a sub-Antarctic island located in the southwest Atlantic (Fig. 1). Water temperatures around this area vary from −0.95 °C in winter to 1.75 °C in summer. South Georgia is a highly productive area of the Southern Ocean, therefore it holds large populations of seabirds, marine mammals and it is one of the most important Southern Ocean fishing areas (Collins et al., 2004; Murphy et al., 2007).

The samples were collected from the Scotia Sea, around South Georgia (Fig. 1), in scientific cruises during the austral summer of 2006/07 [on board of the Royal Research Ship (RRS) *James Clark Ross* (JCR)]: cruise JR161 [October−December 2006], 2007/08 - JR177 [December 2007−February 2008], 2008/09 - JR200 [March−April 2009], Fishing Vessel (FV) *Sil* research survey SG13 [13 January 2013]) and RRS JCR cruise JR16003 (December 2016−January 2017), 2016/17.

On board the RRS *JCR,* samples were collected using an 8 or 25 m^2 mouth-opening Rectangular Midwater Trawl [RMT8 - mesh size reducing from 4.5 mm to 2.5 mm in the cod end; RMT25 - mesh size reducing from 8 mm to 4.5 mm in the cod end (Roe and Shale, 1979)]. The nets were rigged with two opening/closing nets that could be remotely opened and closed at different depths. Samples were collected from 1000 m deep to surface. Cephalopods were identified (Nesis, 1987; Xavier and Cherel, 2009), measured and weighed on board. Samples were preserved individually in separate ziplock bags at −20 °C for later laboratory analyses.

The samples collected by FV *Sil* were obtained from bottom trawls using an FP120 trawl net with a standard steel bobbin rig, conducted at tow speeds between 3.1 and 4.1 knots over a distance of between 1.25 and 2.1 nautical miles, dependent on the prevailing sea conditions and bottom topography. Whenever possible, samples were identified on board but, in some cases, identification was not possible. In each case, individuals were frozen at −20 °C for later laboratory processing.

2.2. Laboratory procedures

All samples were checked for identification [using cephalopod beaks to confirm identification where there was any doubt (Xavier
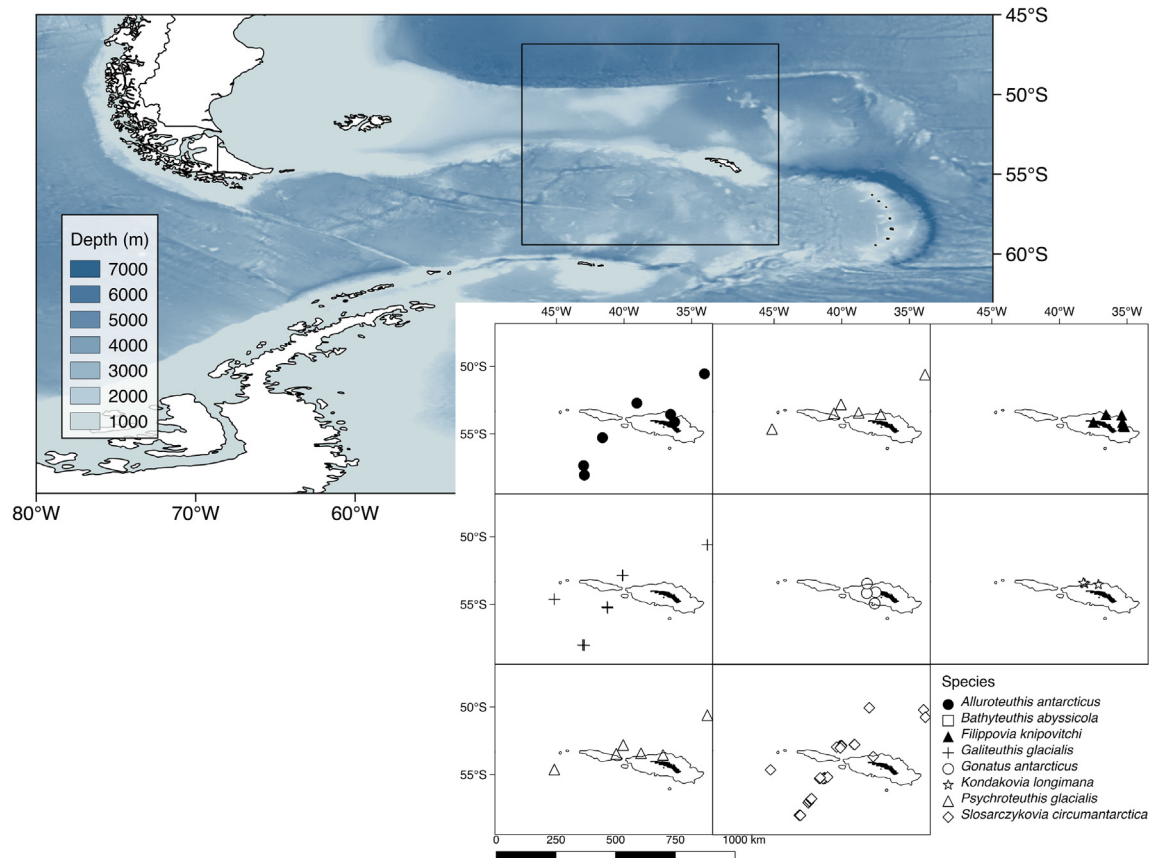
Fig. 1. Sampling sites of species captured around South Georgia across all sampling years. Lines represent 1000 m isobath.

and Cherel, 2009)], measured and weighed again. When the measurement of the mantle length (ML) of the individual was not possible, allometric equations were used, based on beak size (Xavier and Cherel, 2009). An effort was made to collect samples of muscle, gills and digestive gland in all individuals, although the digestive gland was destroyed in some specimens.

After being dissected, the collected tissues were frozen in sterilised decontaminated plastic vials and freeze-dried for at least 48 h. Dried tissues were homogenized and analysed for total mercury by thermal decomposition atomic absorption spectrometry with gold amalgamation, using a LECO AMA-254 (Advanced mercury analyser) following Coelho et al. (2008). Organic mercury was determined through digestion with a mixture of 18% potassium bromide (KBr) in 5% sulphuric acid (H_2SO_4), followed by extraction of organic mercury into toluene (C_7H_8) and back-extraction with an aqueous solution of thiosulphate ($Na_2S_2O_3$), as described in Válega et al. (2006). Where there was low individual mass (less than 200 mg), samples were aggregated in groups of the same species, with similar sizes and collected from the same location. Analytical quality control was performed using three certified reference materials (CRM): NIST 2976 mussel tissue, ERM-CE278K mussel tissues and TORT-3 lobster hepatopancreas. The obtained values (mean ± SD) for the whole of the CRM analyses ranged from 82 to 105% (NIST 2976: 85 ± 7%; ERM-CE278K: 92 ± 5%; TORT-3: 93 ± 8%). Analyses were performed in duplicate, and the coefficient of variation between replicates never exceeded 10%. TORT-3 was also used to validate organic mercury analyses, with an extraction efficiency of 95 ± 10%. The limit of detection for this analytical method was 0.01 ng of absolute mercury. All concentration data are expressed as a function of dry weight (dw).

2.3. Statistical analysis

All analyses were performed using the R software version 3.4.2 (R Core Team, 2017). Correlations were determined between the mercury concentration and the mantle length. Hg levels were tested for normality using Shappiro-Wilk normality test and homogeneity using a Bartlett's test. Friedman tests were used to compare Hg values between the different tissues (muscle, gills, digestive gland) and a Wilcoxon Signed Ranks Test for the tissues of *G. antarcticus* (muscle, gills). Wilcoxon rank test and Kruskal−Wallis test were used to compare Hg in muscle tissue between different years, followed by Dunn's test multiple comparison test.

All values are presented as mean ± SD. The significance level for statistical analyses was always set at $\alpha = 5\%$.

3. Results

3.1. Total mercury concentrations in muscle of Antarctic squid

Overall, total mercury values in muscle were: 63 ± 53 ng g^{-1} in *A. antarcticus* (family Neoteuthidae), 110 ± 40 ng g^{-1} in *B. abyssicola* (family Bathyteuthidae), 100 ± 80 ng g^{-1} in *G. glacialis* (family Cranchiidae), 24 ± 21 ng g^{-1} in *P. glacialis* (family Psychroteuthidae) and 20 ± 20 ng g^{-1} in *S. circumantarctica* (family Brachioteuthidae).

Total mercury was analysed in the mantle muscle of squid species where there were sufficient individuals over a range of sizes to evaluate the pattern of bioaccumulation with size as a proxy for age (Fig. 2). To avoid the effect of sampling year, specimens were selected from the year with higher number of individual (2007/08
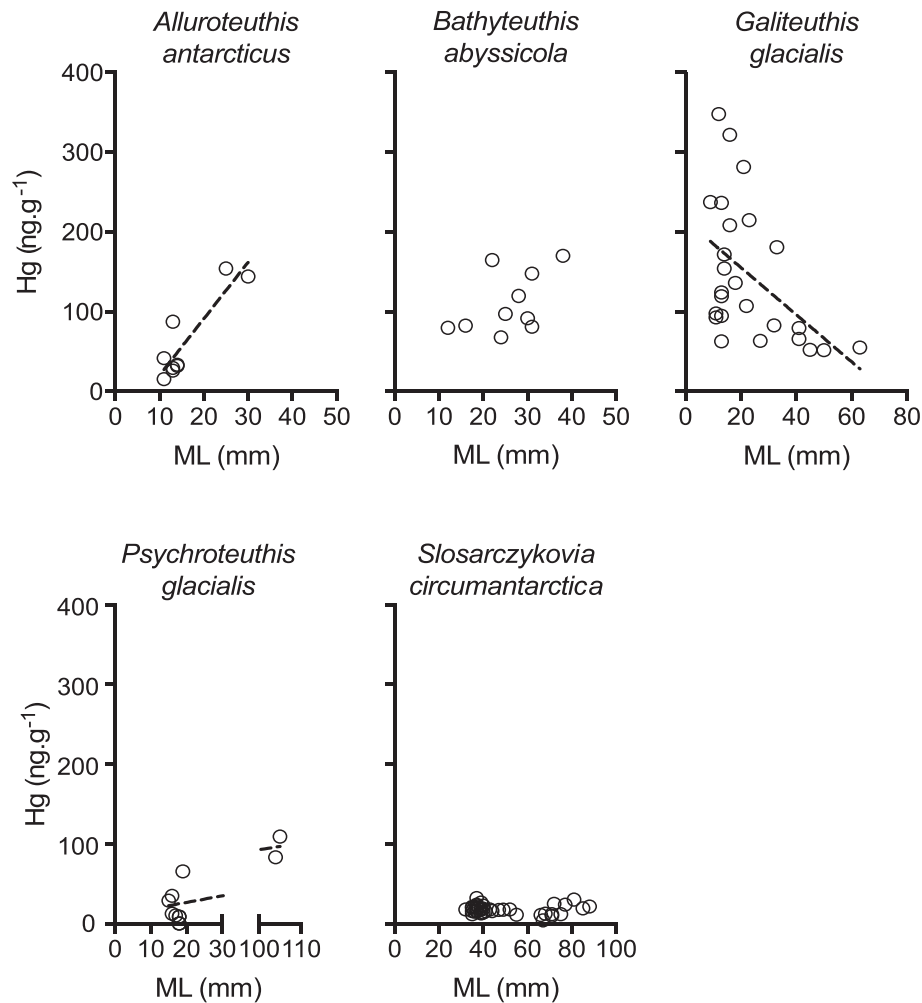
Fig. 2. Total mercury concentration (ng g^{-1} dw) versus mantle length (ML; mm) in individual Antarctic squid collected around South Georgia in the austral summer of 2007/08 (except *Psychroteuthis glacialis* 2016/17). Regression equations are given in the text.

for *B. abyssicola, G. glacialis, A. antarcticus* and *S. circumantarctica*; 2016/17 for *P. glacialis*). Three patterns were noted: 1) an increasing concentration of total mercury with size [*A. antarcticus* (Y = 7.074*X − 50.4, $R^2 = 0.80$, $F_{1, 7} = 28.78$, p = 0.001) and *P. glacialis* (Y = 0.8259*X + 10.1, $R^2 = 0.74$, $F_{1, 7} = 19.68$, p = 0.003)]; 2) no change in total mercury concentration with size [*B. abyssicola* (Y = 2.432*X + 47.83, $R^2 = 0.24$, $F_{1, 8} = 2.56$, p = 0.15) and *S. circumantarctica* (Y = −0.03111*X + 19.33, $R^2 = 0.009$, $F_{1, 38} = 0.33$, p = 0.56)]; 3) a decrease in total mercury concentration with size [i.e. *G. glacialis* (Y = −2.958*X + 214.8, $R^2 = 0.2426$, $F_{1, 23} = 7.366$, p = 0.012)].

3.2. Differential tissue accumulation of total mercury in Antarctic squid

Analysis of mercury accumulation in different tissues was possible in four species (*A. antarcticus, F. knipovitchi, K. longimana* and *P. glacialis*). *F. knipovitchi* was the only species where there were significant differences between tissues (Friedman test: $\chi^2 (2) = 7.6$, p = 0.024), with the digestive gland having a concentration 3 times higher than the muscle and gills. In the other three species, no statistical differences were observed between the mercury concentrations of the three tissues investigated (Friedman test: $\chi^2 (2) = 3$, p = 0.5; $\chi^2 (2) = 3$, p = 0.5; $\chi^2 (2) = 3$, p = 0.5; *A. antarcticus, K. longimana* and *P. glacialis*, respectively; Fig. 3). For *G. antarcticus*,

only the muscle and the gills were analysed, and no differences between these two tissues were found (Wilcoxon Signed Ranks Test; Z = −1.826, p = 0.125).

3.3. Temporal trends of total mercury concentrations in muscle of Antarctic squid

Over the 10 year study period, there was a suggestive decreasing trend in mercury concentrations in the analysed species (Fig. 4).

There were no differences in body size between the years for all the analysed species (*A. antarcticus,* Mann Whitney test, U = 9, p = 0.889; *B. abyssicola,* Kruskal–Wallis H = 1.754, p = 0.442; *G. glacialis,* Kruskal–Wallis H = 10.84, p = 0.442; *P. glacialis,* Mann Whitney test, U = 4.5, p = 0.8; *S. circumantarctica,* Kruskal–Wallis H = 65.46, p = 0.156). *A. antarcticus* and *P. glacialis* were each only caught in two sampling years. For *A. antarcticus*, total mercury was similar between the years 2007/08 (90 ± 60 ng g^{-1}) and 2008/09 (30 ± 3 ng g^{-1}; Wilcoxon rank test; W = 14, p = 0.19), as was *P. glacialis* between 2008/09 (10 ± 2 ng g^{-1}) and 2016/17 (40 ± 20 ng g^{-1}; Wilcoxon rank test; W = 10, p = 0.057). *B. abyssicola* were caught in three years, 2006/07 (150 ± 20 ng g^{-1}), 2007/08 (80 ± 10 ng g^{-1}) and 2016/17 (80 ± 1 ng g^{-1}), with individuals from 2006/07 having statistically higher concentrations of mercury than 2007/08 and 2016/17 (Kruskal–Wallis H = 6.709, p = 0.013; Fig. 4). There were no differences between samples
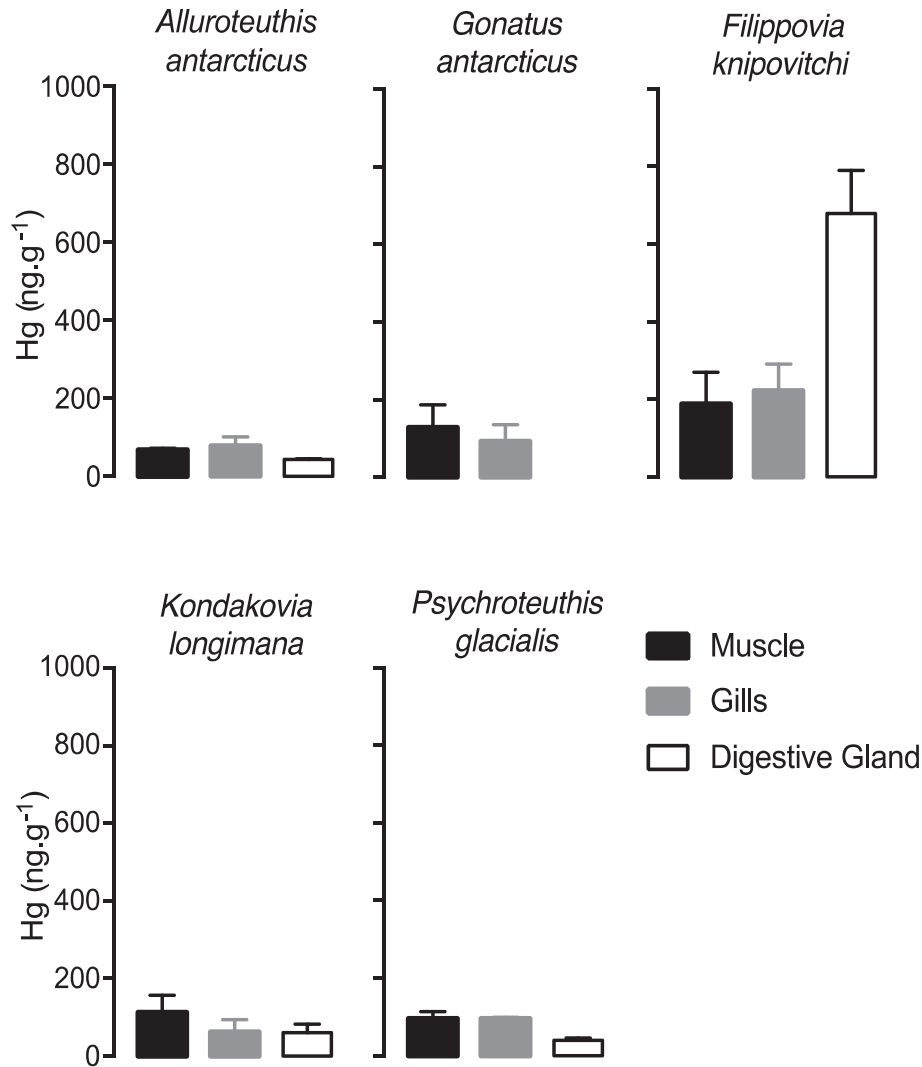
Fig. 3. Total mercury concentrations (Mean $\pm$ SD, ng g^{-1} dw) in different tissues (muscle, gills and digestive gland) of Antarctic squid collected around South Georgia in the austral summer of 2012/2013. No digestive gland tissue was available for G. *antarcticus*.

collected on the other two years. *G. glacialis* were caught on the four sampling years, 2006/07 (100 ± 60 ng g^{-1}), 20007/08 (150 ± 90 ng g^{-1}), 2008/09 (20 ± 10 ng g^{-1}) and 2016/17 (20 ± 7 ng g^{-1}). Mercury concentrations for this species were similar between the years 2006/07 and 2007/08 (Dunn's test Q = -8.77, p = 0.52) and also similar between the years 2008/09 and 2016/17 (Dunn's test Q = 0.057, p > 0.99), but were different between the two similarity groups (2006/07, 2007/08 higher than 2008/09, 2016/17 (Kruskal–Wallis H = 28.69, p < 0.001). *S. circumantarctica* was also caught in all the sampling years: 2006/07 (70 ± 10 ng g^{-1}), 2007/08 (20 ± 6 ng g^{-1}), 2008/09 (10 ± 6 ng g^{-1}) and 2016/17 (10 ± 7 ng g^{-1}); individuals from 2006/07 had statistical higher concentration than the other 3 sampling years (Kruskal–Wallis H = 60.08, p < 0.001).

3.4. Organic mercury concentrations and proportions in Antarctic squid

Regarding the percentage of organic mercury relative to total mercury in the different tissues of the five squid species sampled in 2013 (SG13 samples; Table 1), muscle had the highest values (67%–97%) in all the analysed species, followed by the digestive gland

(22%–38%) and the gills (9%–19%).

Organic mercury concentrations were also analysed in the muscle of three species (*B. abyssicola*, *G. glacialis* and *S. circumantarctica*, Table 2) across sampling years. Concentrations varied from 2 ng g^{-1} to 84 ng g^{-1}, constituting between 25% and 77% of total mercury. *S. circumantarctica* was the only species where organic mercury was lower than 50% (40% in 2007/08; 47% in 2008/09; 25% in 2016/17). Although organic mercury proportions differed significantly between species, there were no statistical differences between years.

4. Discussion

While some data exist on mercury concentrations in Antarctic cephalopods (Anderson et al., 2009; Cipro et al., 2018; McArthur et al., 2003), to the best of our knowledge, this is the first study to measure total and organic mercury concentrations, and to determine the percentage of organic mercury relative to total mercury in different tissues, across a range of Southern Ocean squid species, along a temporal scale.
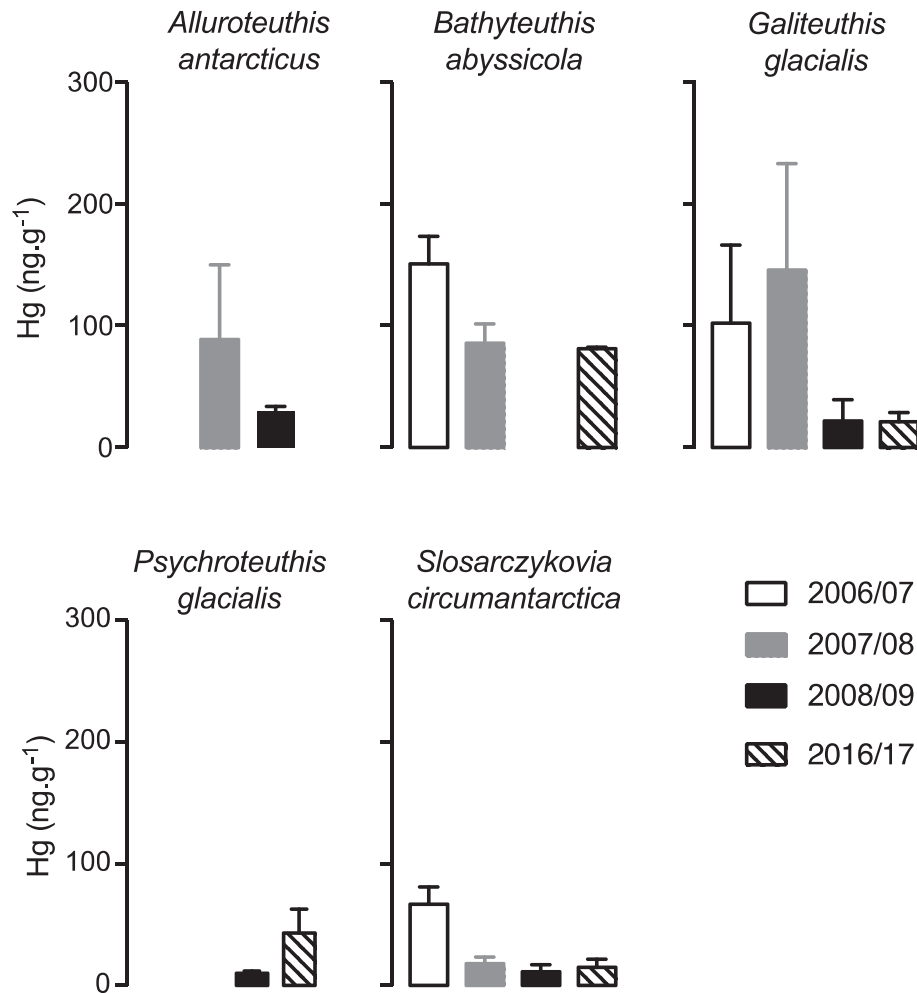
Fig. 4. Total mercury concentrations (Mean ± SD, ng g^{-1} dw) in the muscles of Antarctic squid from the Southern Ocean collected around South Georgia in the austral summers of 2006/07, 2007/08, 2008/09 and 2016/17.

Table 1
Total mercury (THg) and organic mercury (OHg) concentrations (ng g^{-1} dw) and percentage of OHg relative to THg in different tissues (digestive gland, gills and muscle) of Antarctic squid collected around South Georgia in the austral summer of 2013/14.

Species	Tissue	THg	OHg	%OHg
Alluroteuthis antarcticus	Digestive gland	42 ± 2	16 ± 3	38%
	Gills	60 ± 23	11 ± 2	18%
	Muscle	65 ± 4	52 ± 12	80%
Filippovia knipovitchi	Digestive gland	643 ± 98	144 ± 52	22%
	Gills	306 ± 68	57 ± 14	19%
	Muscle	79 ± 28	53 ± 9	67%
Gonatus antarcticus	Gills	89 ± 33	8 ± 2	10%
	Muscle	162 ± 58	158 ± 29	97%
Kondakovia longimana	Digestive gland	45 ± 21	11 ± 3	25%
	Gills	75 ± 32	9 ± 4	12%
	Muscle	82 ± 23	78 ± 14	95%
Psychroteuthis glacialis	Digestive gland	25 ± 6	6 ± 3	24%
	Gills	99 ± 3	9 ± 4	9%
	Muscle	83 ± 18	62 ± 22	74%

Table 2
Total mercury (THg) and organic mercury (OHg) concentrations (ng g^{-1} dw) and percentage of OHg relative to THg in the muscles of Antarctic squid collected around South Georgia in the austral summers of 2006/07, 2007/08, 2008/09 and 2015/16.

Species	n	Year	THg	OHg	%OHg
Bathyteuthis abyssicola	3	2006/07	103 ± 14	71 ± 12	69%
Galiteuthis glacialis	4	2006/07	40 ± 8	30 ± 5	75%
	4	2007/08	110 ± 26	60 ± 13	55%
	2	2008/09	26 ± 6	20 ± 5	77%
	2	2016/17	43 ± 2	24 ± 2	56%
Slosarczykovia circumantarctica	6	2007/08	20 ± 12	8 ± 5	40%
	4	2008/09	15 ± 5	7 + 3	47%
	2	2016/17	16 ± 7	4 ± 2	25%

4.1. Total mercury concentrations according to size in Antarctic squid

Biological and environmental factors such as size, sex, prey preferences and habitat are drivers for mercury concentrations in cephalopods (Bustamante et al., 2006; Monteiro et al., 1992; Storelli

and Marcotrigiano, 1999). When looking into the relationship between size and contamination level in squid from South Georgia (Fig. 3), it is possible to identify three different patterns: 1) *A. antarcticus* and *P. glacialis* had a positive correlation between contamination level and size, 2) The correlation for *G. glacialis* was negative, 3) Mercury concentration did not appear to be related to size in *B. abyssicola* or *S. circumantarctica*.

Significant variation and opposite patterns between size and mercury concentrations in cephalopods have been reported previously. Mercury concentrations increased with size in the veined squid *Loligo forbesi* (Chouvelon et al., 2011; Monteiro et al., 1992;

Pierce et al., 2008), the neon flying squid *Ommastrephes bartramii* (Monteiro et al., 1992), the orange-back flying squid *Sthenoteuthis pteropus* (Lischka et al., 2018), the common octopus *Octopus vulgaris* (Rjeibi et al., 2014), the curled octopus *Eledone cirrhosa* (Rossi et al., 1993), the common cuttlefish *Sepia officinalis* and the European squid *Loligo vulgaris* (Chouvelon et al., 2011). However, in *O. vulgaris* (Raimundo et al., 2004, 2010a) and the greater hooked *Onykia ingens* (McArthur et al., 2003), no relationships were found, while in the spider octopus *Octopus salutii* (Storelli and Marcotrigiano, 1999), *O. vulgaris* and *Loligo vulgaris* (Rjeibi et al., 2014) there were negative relationships. Hence, the effect of size on mercury bioaccumulation in cephalopods is variable (Penicaud et al., 2017), and is likely to be sensitive to fluctuating environmental conditions (see below).

4.2. Tissue allocation of mercury in Antarctic squid

For four species (*A. antarcticus, G. antarcticus K. longimana* and *P. glacialis*), there were no differences in total mercury concentrations between tissues. However, the fraction of organic mercury was always higher in muscle (67%–97%) than in the digestive gland (22%–38%) and gills (9%–19%). While presently the reasons for the mercury distribution patterns are still a matter of debate, the partitioning of total mercury among tissues is consistent with previous studies on oceanic squid [Cranchidae, Histioteuthidae and Ommastrephidae (Bustamante et al., 2006)], suggesting some degree of equilibrium between contaminant accumulation and excretion rates, as well as efficient redistribution of total mercury between tissues. In most studies of cephalopod species however, the digestive gland had higher total mercury concentrations than other tissues (Bustamante et al., 2006; Pierce et al., 2008; Raimundo et al., 2010b), as we observed in *F. knipovitchi* (Fig. 4). The high rates of absorption and assimilation of trace metals in the digestive gland (Bustamante et al., 2002; Miramand and Bentley, 1992; Raimundo et al., 2004) make this organ a major pathway of incorporation for contaminants into cephalopods (Bustamante et al., 2002), and reflects the predominant role of the dietary pathway for mercury bioaccumulation. The lower concentrations of organic mercury in the digestive gland is in accordance with the detoxification of organic mercury by demethylation that is suspected to occur in this organ (Bustamante et al., 2006; Penicaud et al., 2017). As this form of mercury is incorporated from the diet, a fraction is likely to be demethylated in the digestive gland, to allow its excretion (Lacoue-Labarthe et al., 2009), and a portion will be circulated, trapped and stored in muscle tissues. Because organic mercury has a strong affinity for the sulphydryl groups of proteins, its accumulation in muscle tissues is favoured (Bloom, 1992; Leaner and Mason, 2004). In fish muscles, for instance, it may be tightly bound by carbon-mercury and sulphydryl linkages (Ruelas-Inzunza et al., 2003). As muscles comprise the highest mass fraction of cephalopods, and given the known affinity of organic mercury to muscular proteins (Bloom, 1992), high concentrations and proportions of organic mercury are to be expected in this tissue. The present results also highlight the high bioavailability of mercury within the Southern Ocean food web (Chouvelon et al., 2012; Clarkson, 1992; Dehn et al., 2006).

Concentrations and proportions of organic mercury also show a considerable variation between species. The high intra-species variation may be explained by the dietary ontogenetic shift that occurs in cephalopods (Xavier et al., 2018) when they change from feeding on small crustaceans [lower percentage of organic mercury (Seco et al., 2019)] to prey at higher trophic levels (higher percentage of organic mercury (Chouvelon et al., 2011)), however this can not be applied to *G. glacialis,* as mercury in this species decreases in bigger individuals. Similarly, interspecific differences are

probably due to variations in diet (Boyle and Rodhouse, 2005), although the trophic ecology of these cephalopods is still poorly known (Collins and Rodhouse, 2006).

4.3. Temporal trends of mercury concentrations in Antarctic squid

Our study analysed samples obtained over a 10 year period (between 2006/07 and 2016/17), although only two species were captured in all sampling seasons (*G. glacialis, S. circumantarctica*) and the number of individuals captured for some species was relatively low despite the sustained sampling effort. Squid have a highly developed sensory system and can swim fast, while sampling nets are relatively small and slow, so most adult squid can avoid capture (Clarke, 1977; Xavier et al., 2002).

Squid are considered an r-selected species (Pianka, 1970): they have a fast growth rate, are semelparous (reproduce only once) and are short lived (~< 1–2 years) (Arkhipkin, 2004; Boyle and Rodhouse, 2005; Xavier et al., 2018), although, some large squid species, like *K. longimana* may live longer (Jarre et al., 1991; Lynnes and Rodhouse, 2002). These characteristics make them responsive bioindicators with which would be possible to monitor trends in mercury concentrations over time, as they will reflect rapidly any changes in the bioavailability of this contaminant.

Data from the squid species *A. antarcticus*, *B abyssicola*, *G. glacialis* and *S. circumantarctica* show a decreasing trend of mercury concentration along time, although further monitoring is required to confirm this pattern. This suggestive pattern of declining concentrations of mercury in the majority of species is consistent with the decreasing trend of atmospheric mercury over the last decade (Soerensen et al., 2012) as a consequence of the reduction of worldwide anthropogenic emissions of mercury (Streets et al., 2017; Zhang et al., 2016). The decrease of mercury in the global atmosphere could also mean a reduction in mercury deposition levels in our study area. Comparing our results with a previous study in the same region (Anderson et al., 2009) who sampled in 2001/02 and analysed specimens with atomic fluorescence spectrophotometry, a general reduction in the concentration of mercury can also be observed: *G. glacialis* had a mercury concentration in 2001/02 (230 ± 70 ng g^{-1}) (Anderson et al., 2009) that was more than twice as high as our results from 2006/07 (100 ± 60 ng g^{-1}), 1.5 times when compared with 2007/08 (150 ± 90 ng g^{-1}) and more than ten times when compared with 2008/09 or 2016/17 (20 ± 10 ng g^{-1}; 20 ± 7 ng g^{-1}); in *G. antarcticus* (600 ± 2 ng g^{-1}) mercury concentrations were 5 times higher in 2001/02 than our results for 2013. *P. glacialis* had concentration 4 times higher (180 ± 110 ng g^{-1}) in 2001/02 than our observations in 2008/09. In the family Onychoteuthidae, the concentrations of mercury in 2001/02 (100 ± 20 ng g^{-1} in *F. knipovitchi;* 160 ± 90 ng g^{-1} in *K. longimana*) were similar to our results for 2013.

The pattern of mercury bioaccumulation in species is influenced by specific traits such as dietary preference, ingestion, excretion, and growth rate. Prevailing environmental conditions may also enhance or reduce contaminant bioavailability to cephalopods. Habitat use has a major effect on mercury accumulation in organisms, as sites contaminated by mercury will likely have higher bioavailability of this toxic element. All of our samples were collected in the Scotia Sea, around South Georgia (Fig. 1), which is known to be a fairly stable ecosystem, which should mean lower mercury variation between sampling sites.

The majority of our study species have a wide range of vertical distributions: the depth of occurrence of *A. antarcticus* is normally at 800–900 m (Rodhouse, 1988), *G. glacialis*, at 600–1000 m (Roper and Young, 1975), *G. antarcticus,* at 250–928 m (Collins et al., 2004; Roper et al., 1984), *F. knipovitchi*, at 480–760 m (Collins et al., 2004), *K. longimana,* at 300–900 m (Collins et al., 2004; Xavier et al.,

2002), *P. glacialis,* at 275−928 m (Collins et al., 2004; Xavier et al., 2002) and *S. circumantarctica,* at 487−928 m (Collins et al., 2004). An outlier amongst these is *B. abyssicola*, which is a deep-sea squid and occurs at a depth of 1000−1500 m (Roper and Young, 1975). Higher mercury levels were expected in species from deep habitats as increasing depth raises mercury concentrations in fish (Choy et al., 2009; Le Bourg et al., 2019; Monteiro et al., 1996). However, that was not the case in our study, as mercury concentrations in the tissues of the deep-sea squid *B. abyssicola* were within the same range as the majority of other analysed species (see Fig. 4). The lack of difference between species may be explained by the frequent diel vertical migration associated with cephalopods (Norman et al., 2014) in order to feed and to avoid predation. The differences in mercury concentrations between years may be partially explained by shifts in the abundance and contaminant loads of prey items (Chouvelon et al., 2011; Paiva et al., 2008). However, there is still a lack of data on the diets of the studied species (Collins and Rodhouse, 2006; Xavier et al., 2018; Xavier and Cherel, 2009) and it is presently not possible to evaluate the effect of diet on mercury bioaccumulation in our study species.

Our results suggest a decreasing trend of total mercury concentration in most of the South Georgia squid species analysed over the last decade, with a stable proportion of organic mercury. Considering that cephalopods are a major link between primary consumer and top predators, these changes possibly reflect a drop in mercury bioavailability in lower trophic levels and suggests that mercury intake by squid predators may have decreased.

Acknowledgments

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