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Mercury in the ecosystem of Admiralty Bay, King George Island, Antarctica: Occurrence and trophic distribution



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ABSTRACT

Mercury (Hg) can reach the environment through natural and human-related sources, threatening ecosystems all over the planet due to its well known deleterious effects. Therefore, Antarctic trophic webs, despite being relatively isolated, are not exempt of its influence. To evaluate Hg concentrations in an Antarctic ecosystem, different tissues from 2 species of invertebrates, 2 of fish, 8 of birds, 4 of pinnipeds and at least 5 of vegetation were investigated ($n = 176$). For animals, values ranged from 0.018 to 48.7 $\mu\text{g g}^{-1}$ dw (whole Antarctic krill and Antarctic Fur Seal liver). They were generally correlated to trophic position (assessed by $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$) but also to cephalopods and myctophids consumption. For vegetation, values ranged from 0.014 to 0.227 $\mu\text{g g}^{-1}$ dw (*Colobanthus quitensis* and an unidentified lichen), with lichens presenting significantly higher values than mosses, likely due to year-round exposure and absorption of animal derived organic matter, as hypothesized by literature.

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Mercury (Hg), besides its natural sources, can reach the environment through human-related activities, such as mining and the burning of fossil fuels (Fitzgerald et al., 1998, 2007; Selin, 2009). Indeed, non-natural sources play a major role in Hg deposition, estimated to occur in a current rate three to five times higher than its pre-industrial times (Selin, 2009).

In the open ocean the main Hg source is atmospheric deposition of inorganic Hg (II) by both wet and dry processes (e.g. Fitzgerald et al., 2007). Over 80% of the Hg deposited in marine ecosystems is reemitted to the atmosphere (as Hg(0) predominantly, but some as $(\text{CH}_3)_2\text{Hg}$), increasing the residence time of Hg cycling through the reservoirs of the surface biosphere (Driscoll et al., 2013). Once in the water column, inorganic Hg can be methylated by anaerobic microorganisms, such as sulphate-reducing and iron-reducing bacteria (e.g. Hsu-Kim et al., 2013). The produced Me-Hg (mono-methyl-Hg) is then easily assimilated by the biota and biomagnifies up the food web (e.g. Morel et al., 1998). Because microorganisms activity is importantly influenced by temperature, springtime conversion of Hg into Me-Hg may provide an important environmental pathway for its introduction into the biosphere in a time of the year when biota are preparing for peak summer-time activity (Schroeder et al., 1998).

In the Southern Hemisphere, high concentrations of atmospheric Hg have been observed close to the Antarctic continent when compared to

lower latitudes (Soerensen et al., 2010). Antarctica, in spite of being the most isolated continent on Earth, is far from exempt of the input of several sorts of contaminants, including Persistent Organic Pollutants such as PCBs or organochlorine pesticides, as reported in literature (e.g. Bargagli et al., 2007; Corsolini, 2009). Antarctica is thus contaminated by anthropogenic Hg which can affect the fitness of organisms and have consequences at the population level as shown on polar skuas (Goutte et al., 2014).

Investigating contaminants such as Hg in trophic webs can be greatly improved by using stable isotopes of carbon and nitrogen as an ecological tool to shed light, respectively, the carbon sources exploited by consumers and their trophic position (Lesage et al., 2002). Previous studies on Arctic ecosystems have already shown that Hg analyses coupled to $\delta^{15}\text{N}$ (e.g. Atwell et al., 1998) or both $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ (e.g. Aubail et al., 2011) allows a better understanding of Hg biomagnification pathways and moreover, the influence of feeding ecology factors such as change in feeding sites, gender or age class trophic segregation, i.e., trophic position as a whole.

Since the late 70s, organic contaminants have received interest in King George Island (Lukowski, 1978) but with a relative lack of data for stable isotopes, with only a small quantity of studies taking SIA into account (e.g. Corbisier et al., 2004 and Majer et al., 2014 for benthic invertebrates; Cipro et al., 2011 for vegetation), particularly for upper vertebrates (e.g. Cipro et al., 2012). Also, a lack of Hg data (e.g. dos Santos et al., 2006) is obvious for the organisms from this area. In this context, this study is the first, to the best of our knowledge, coupling Hg and isotopic data for organisms from King George Island.

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Taking all of that into account, the study aims at assessing Hg concentrations in the light of SI ratios; at raising hypotheses on how these results are influenced by one another and also by ecological factors such as diet, distribution, exposure pathways and so on; and finally, at comparing the results with those from literature, investigating the factors that might play important roles in these eventual differences.

Sample collection took place in King George Island (62°05'S, 058°23'W), the largest one of the South Shetland Islands, separated from the northern portion of the Antarctic Peninsula by the Bransfield Strait. The number of samples of each species, tissue and analysis is given in Table 1. All samples were kept in previously combusted (at least 420 °C for at least 4 h) containers, frozen upon arrival (−20 °C) at the Brazilian Antarctic Station and kept frozen until freeze-drying in the LabQOM (University of São Paulo, Brazil). Hg and stable isotope analyses (SIA) analyses were performed at the University of La Rochelle (France). Briefly, bird eggs and tissues, fish and invertebrate samples were collected according to Cipro et al. (2010, 2013). Limpets (Antarctic limpet, *Nacella concinna*) were manually collected (2004–05 summer) in the intertidal zone. Fish (marbled rockcod, *Notothenia rossii* and black rockcod *N. coriiceps*) were collected (2006–07 summer) by mid-water nets or by line and hook. Only unhatched bird eggs (skua, *Catharacta* sp., identified only until the genus level; kelp gull, *Larus dominicanus*; and Antarctic tern, *Sterna vittata*) were collected (2004–05 and 2005–06 summers) so as not to interfere with breeding success. Bird livers (cape petrel, *Daption capense* and giant petrel, *Macronectes giganteus* in addition to the previous species) were collected (from 2004–05 up to 2007–08 summers) only from already dead animals with no evident signs of disease, decomposition or emaciation. No further attempt to determine the cause of death was performed. Pinnipeds samples were collected according to Cipro et al. (2012), which already contains the SIA

data: samples of the Weddell seal, *Leptonychotes weddellii*; Antarctic fur seal, *Arctocephalus gazella*; crabeater seal, *Lobodon carcinophagus*; and southern elephant seal, *Mirounga leonina*, were collected during the austral summers of 2004/05 and 2005/06, in a fully opportunistic manner, i.e., only from animals found already dead, with no signs of degradation. Finally, vegetation samples were collected according to Cipro et al. (2011), which contains also the SIA data: the angiosperm *Colobanthus quitensis*, the mosses *Brachythecium* sp., *Syntrichia princeps* and *Sanionia uncinata* and the lichens *Usnea aurantiaco-atra* and *Usnea antarctica* were collected from early December 2004 to early January 2005.

Prior to stable isotope and Hg analyses, samples were lyophilized and ground to obtain a fine powder and stored in plastic vials.

Mercury analyses were carried out with an Automatic Mercury Analyser spectrophotometer, ALTEC AMA 254, which does not require an acid digestion of the samples. Aliquots ranging from 10 to 50 mg of freeze-dried sample (with no delipidation whatsoever) were directly analysed after being inserted in the oven of the apparatus. After drying, the samples were heated under an oxygen atmosphere for 3 min, and the Hg liberated and subsequently amalgamated on an Au-net. The net was then heated to liberate the collected Hg, which was measured by Atomic Absorption Spectrometry (AAS). Accuracy and reproducibility of the method were tested using dogfish liver (DOLT-2) and muscle (DORM-2) and lobster hepatopancreas (TORT-2) (National Research Council, Canada) reference standards. Standard and blanks were analysed along with each set of samples, and recoveries of the certified values and recoveries of the metals ranged from 93 to 109%. Measurements were also validated by IAEA inter-calibration exercises (Coquery et al., 2001). Concentrations are expressed in dry weight in order to compensate eventual moisture loss during freezing and to

Table 1
SIA ($\delta^{13}\text{C}$ and $\delta^{15}\text{N}$, in ‰) and Hg (in $\mu\text{g g}^{-1}$, dry weight), with its respective standard deviations (σ).

Species	Tissue	n	$\delta^{13}\text{C}$	σ	$\delta^{15}\text{N}$	σ	Hg	σ
<i>Euphausia superba</i>	Whole	4	−25.66	0.69	4.51	0.53	0.018	0.005
<i>Nacella concinna</i>	Soft part	8	−16.10	1.74	7.27	1.23	0.037	0.015
<i>Notothenia coriiceps</i>	Muscle	1	−20.06	–	10.80	–	0.093	–
<i>N. rossii</i>	Muscle	28	−21.53	1.09	10.21	0.69	0.077	0.027
<i>Catharacta</i> sp.	Egg	4	−24.34	0.14	11.13	0.39	2.520	1.709
	Liver	3	−24.08	0.47	12.11	0.44	5.136	5.701
<i>Daption capense</i>	Liver	2	−25.76	–	10.45	–	6.965	–
<i>Larus dominicanus</i>	Egg	3	−23.36	2.29	9.13	0.68	0.195	0.067
	Liver	3	−23.42	2.69	11.64	3.74	11.02	16.82
<i>Macronectes giganteus</i>	Liver	3	−23.46	1.97	12.38	2.77	25.47	31.63
<i>Pygoscelis adeliae</i>	Egg	3	−25.27	1.06	9.66	0.53	0.075	0.049
	Liver	5	−26.11	1.31	8.67	0.96	1.742	3.527
<i>P. antarctica</i>	Egg	26	−25.52	0.55	9.99	0.29	0.599	0.273
	Liver	16	−25.38	1.87	8.93	1.46	1.906	1.265
<i>P. papua</i>	Egg	9	−25.96	0.53	9.27	0.24	0.133	0.037
	Liver	16	−25.88	0.57	8.99	0.68	0.369	0.275
<i>Sterna vittata</i>	Egg	1	−23.32	–	9.39	–	0.664	–
<i>Arctocephalus gazella</i>	Liver	1	−25.01	–	11.13	–	48.71	–
	Muscle	2	−24.19	0.43	9.09	1.07	0.091	0.049
	Skin	3	−21.62	–	10.66	–	0.028	–
<i>Leptonychotes weddellii</i>	Kidney	1	−25.55	–	8.02	–	0.075	–
	Liver	1	−24.31	–	12.54	–	0.286	–
	Muscle	2	−24.02	1.00	10.71	3.90	0.112	0.005
	Skin	2	−23.81	–	14.73	–	0.260	–
<i>Lobodon carcinophagus</i>	Muscle	1	−22.68	–	11.26	–	0.124	–
	Skin	2	−23.32	–	11.84	–	0.191	–
<i>Mirounga leonina</i>	Liver	2	−23.71	–	10.21	–	25.68	–
	Muscle	1	−23.09	–	10.22	–	0.613	–
<i>Colobanthus quitensis</i>	Whole	1	−25.31	–	12.79	–	0.014	–
<i>Brachythecium</i> sp.	Whole	1	−21.64	–	−0.53	–	0.086	–
<i>Sanionia uncinata</i>	Whole	7	−24.32	0.49	6.40	7.32	0.068	0.026
<i>Syntrichia princeps</i>	Whole	2	−24.40	2.25	4.70	3.35	0.057	0.030
Unidentified moss	Whole	1	−24.10	–	11.77	–	0.100	–
<i>Usnea antarctica</i>	Whole	3	−21.13	–	−6.20	–	0.152	0.032
<i>U. aurantiaco-atra</i>	Whole	5	−19.33	1.07	−1.29	5.07	0.132	0.037
<i>Usnea</i> sp.	Whole	1	−20.74	–	−7.67	–	0.210	–
Unidentified lichen	Whole	2	−21.01	–	−1.01	–	0.227	0.115

facilitate comparison between tissues and with other studies. Blanks were analysed at the beginning of each set of samples, and the detection limit of the method was $0.005 \mu\text{g g}^{-1}$ dry mass. Lipid content values and obtaining methods have been previously published (Cipro et al., 2011, 2012, 2013).

For SIA, one aliquot of approx. 100 mg of sample was placed in a test tube with 4 mL of cyclohexane to remove lipids according to Chouvelon et al. (2012a) as lipids are depleted in ^{13}C . The mixture was shaken for an hour, then centrifuged for separation and the liquid discarded. This procedure was repeated as many times as needed until the liquid phase comes out clear. The delipidated pellet was then dried at 50°C for 48 h. Aliquots were analysed using a Thermo Scientific Delta V Advantage, ConFlo IV interface (NoBlank and SmartEA) and Thermo Scientific Flash EA1112 Elemental Analyser. Each injection corresponded to 0.4 ± 0.1 mg for animal sample or 1.0 ± 0.1 mg for vegetation samples, encapsulated in tin cups, and there were no replicates unless C/N ratio were above than 4. In this case, delipidation would be repeated until this condition was met. Pee Dee Belemnite and atmospheric nitrogen were used as standards for calculation of $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$, respectively. Based on replicate measurements of internal laboratory standards, experimental precision is of $\pm 0.15\%$ and $\pm 0.20\%$ for $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$, respectively.

Statistics were performed using Microsoft Excel and Statsoft Statistica 12. Before analyses, data were checked for normality of distribution and homogeneity of variances using Shapiro–Wilk and Brown–Forsythe tests, respectively. Parametrical (Pearson's product-moment correlation, Tukey's HSD/ANOVA) and nonparametrical tests (Spearman's rank correlation, Kruskal–Wallis/ANOVA) followed accordingly. Statistically significant results were set at $\alpha = 0.05$.

Finally, normal distribution and log-link function generalized linear models (GLM) were built as follows: Hg concentrations as the dependent variable, species and tissue as categorical factors and $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values as continuous predictors. Biologically relevant models were constructed incorporating the different variables and their interactions. Continuous variables ($\delta^{13}\text{C}$ and $\delta^{15}\text{N}$) were not significantly correlated in the whole dataset and could be included in the same models. Model selection was based on Akaike's Information Criteria adjusted for small sample sizes (AICc). The model with the lowest AICc value was considered to be the most accurate. Models with AICc values differing by less than 2 have a similar level of support in the data, and the model including the least number of parameters can be regarded as the most accurate, according to the principle of parsimony (Burnham and Anderson, 2002). Overall model support was assessed using Akaike weights (w_i), following (Johnson and Omland, 2004). Residual (R^2 adj) analyses should be restricted to description and not be used in model selection (Burnham and Anderson, 2002). Only models with AICc values differing by less than 10 are presented.

The results for Hg and SIA are presented in Table 1. In regard to intra-specific data for animals, the Antarctic krill, *Euphausia superba*, presented the lowest Hg concentration of the whole animal dataset ($0.018 \pm 0.005 \mu\text{g g}^{-1}$), which comes in agreement with its low trophic level. Reported Hg concentrations for this species from different areas were in the same order of magnitude ranging from 0.016 to $0.041 \mu\text{g g}^{-1}$ along the Antarctic Peninsula (Locarnini and Presley, 1995), averaged $0.033 \pm 0.013 \mu\text{g g}^{-1}$ (after dry weight conversion) in several locations ranging from 63 to 68°S and from 126 to 147°W and also from 107 to 157°E (Yamamoto et al., 1987) and, with values averaging $0.035 \mu\text{g g}^{-1}$ in the same area as in the present study (dos Santos et al., 2006). These authors also bring data for *Nacella concinna*, averaging $0.026 \mu\text{g g}^{-1}$, also consistent with our findings.

The SIA data for the invertebrates show two clearly distinct $\delta^{13}\text{C}$ signatures: a depleted pelagic/oceanic for *E. superba* and a much more enriched for the benthic/coastal *N. concinna*. These values are similar to the ones reported by a comprehensive study in the same region (Corbisier et al., 2004) and are key to understand the mixing of benthic/pelagic organic matter sources throughout the local food web.

This is due to the fact that, besides the feeble trophic enrichment of $\delta^{13}\text{C}$ (DeNiro and Epstein, 1978) and also possible migration (e.g. Cherel et al., 2006), the consumption of both sources will result in a proportionally intermediate signature (e.g. Carravieri et al., 2013), and therefore, clarify feeding ecology for a given species. Even though both these invertebrates are primary consumers, there is an important difference in their $\delta^{15}\text{N}$ (i.e., 2.7%, Table 1). However, the signature of the primary producers they consume presents a similar difference: 0.5‰ for phytoplankton and 4‰ for microphytobenthos in this region (Dunton, 2001), i.e., 3.5‰. Thus, the difference found is indeed a matter of baseline nitrogen signature of the organisms on which they prey and not of trophic level itself (Chouvelon et al., 2012b). According to the significant negative correlation between Hg and $\delta^{13}\text{C}$ ($\rho = -0.828$, paired $n = 6$) in *Nacella concinna*, it seems that some $\delta^{13}\text{C}$ depleted food items might represent a Hg source for this mollusc, since it feeds mainly on the relatively $\delta^{13}\text{C}$ -enriched microphytobenthos (Corbisier et al., 2004), but the diet of this species also comprises microbial films (grazing on the microepiflora), calcareous rhodophytes, seaweed, bryozoans and sessile spirobid polychaetes (Suda et al., 2015). Moreover, a local abiotic compartment may act like an indirect source, similarly as described by Mão de Ferro et al. (2014): limpet samples were collected in the intertidal zone, in most cases next to glaciers, therefore subject to snow melting water, glaciers seasonal melting water (which percolates soil and bedrock before reaching the shore), marine spray, which might all function as Hg sources to the limpets themselves via respiration or to the organisms they feed on. These factors might influence $\delta^{15}\text{N}$ as well, but apparently not in a significant way.

As for the *Notothenia* spp., muscle Hg concentrations for *N. rossii* from Kerguelen waters (averaging $0.255 \mu\text{g g}^{-1}$ dw) were three times higher than the present results (Bustamante et al., 2003). *N. coriiceps* from Laurie Island (South Orkney Islands) presented Hg concentration of $0.200 \mu\text{g g}^{-1}$ dw (converted from the fresh wt assuming a 75% water content) (Moreno et al., 1997). Surprisingly, this value is closer to Bustamante et al. (2003) than to our findings whereas Laurie Island are situated south from the Polar Front and Kerguelen Islands, north. Indeed, predators north of the Polar Front show higher Hg concentrations than those from the southern area. For example, wandering albatrosses foraging South the Polar front are less exposed to Hg than those feeding north (Bustamante et al., 2016; Carravieri et al., 2014b). Lower values compared to our results were also reported for *Notothenia* spp. (with data for whole fish averaging $0.016 \mu\text{g g}^{-1}$; dos Santos et al., 2006), but the analysed individuals are assumed to be juveniles. In adult *N. coriiceps* from Adélie land, muscle Hg concentrations were one order of magnitude higher, i.e., $0.221 \pm 0.085 \mu\text{g g}^{-1}$ dw (Goutte et al., 2015). The $\delta^{13}\text{C}$ signature found in these fish is intermediate in regard to *E. superba* and *N. concinna* which is likely due to the consumption of both oceanic/pelagic and coastal/benthic prey. Moreover, the significant positive correlation between $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ ($\rho = 0.662$) in *N. rossii* confirms that this fish displays ontogenetic changes with growth: as the fish grows, higher trophic level prey is increasingly preferred, specially small fish (Burchett, 1983).

Data for *Catharacta* sp. eggs appears as the most contaminated for this tissue. This might be due to the high trophic level of this species, as shown by elevated $\delta^{15}\text{N}$. Skuas are opportunistic feeders (Reinhardt et al., 2000) and prey on other birds: eggs, chicks or the adults themselves. In locations where skuas have developed a specialisation of their diet such as in Mayes Island in the Kerguelen archipelago, chicks are fed mainly with blue petrels (Mougeot et al., 1998) and they consequently present higher feather Hg levels than adults as a reflect of age-class feeding habits' differences (Carravieri et al., 2014a). Indeed, the parents feed on low trophic level prey in Antarctic waters (Cherel et al., 2002, 2006), with a large proportion of mesopelagic fish (Cherel et al., 2002; Connan et al., 2008). In turn, breeding skuas in King George Island feed mainly on penguins and their eggs, and fish, varying somewhat between these two prey groups for the two occurring species in this location (Peter et al., 1990). Nevertheless, considering feather

data (Carravieri et al., 2014b), as well as diet (Bocher et al., 2003), it is reasonable to assume that this diet would lead to high Hg exposure to skua from the present work. In eggs, the present values ($2.520 \pm 1.709 \mu\text{g g}^{-1} \text{ dw}$) are higher when compared to the $1.61 \pm 1.22 \mu\text{g g}^{-1} \text{ dw}$ for *C. macormicki* eggs from a previous study conducted in Terra Nova Bay (Bargagli et al., 1998a). Even though in seabirds showing gender differences in their foraging strategies such as wandering albatrosses whose females had higher Hg concentrations than males despite the excretion through the eggs because they forage in more contaminated waters (north) than males (south) (Carravieri et al., 2014a), in regard to skuas there seems to be no gender segregation in both Hg concentration and SIA, not even during the breeding season (Bearhop et al., 2000a).

SIA data puts skuas in roughly the same ecological niche as previous literature (Bearhop et al., 2000a, 2000b), particularly when liver is compared to blood, since both these tissues have short and similar turnover rates (Hobson and Clark, 1992). Unpublished data (Cipro et al., personal data) confirms these previous statements, also presenting sub-Antarctic species (from Kerguelen) with higher Hg concentrations than Antarctic ones (from Adélie Land) occupying similar ecological niches, the Cape petrel *Daption capense* presented remarkably high Hg concentrations despite its $\delta^{15}\text{N}$ closer to penguins which showed much lower Hg concentrations. Such high concentrations might be due to two factors. The first one is that its diet is, in a general way, almost entirely composed by oceanic/pelagic prey (Coria et al., 1997) whereas pygoscelids penguins might, in a lesser way, consume some coastal/benthic prey (especially Gentoos penguins, *P. papua*). Pygoscelids also show a reproductive temporal shift, mostly due to their respective distributions, and not necessarily to food availability. These penguins breed in different periods of the austral summer in this area, with Adélies early breeding, followed by Gentoos and then Chinstraps (Trivelpiece et al., 1987). The second one is that most of the Cape petrel diet in the region (Casaux et al., 1998; Coria et al., 1997) is composed by Myctophidae fish, which have higher Hg concentrations compared to epipelagic ones (e.g., Chouvelon et al., 2012a; Monteiro et al., 1996) and species that prey on these resources are therefore more prone to accumulate Hg (Blévin et al., 2013; Carravieri et al., 2014b; Cipro et al., 2014).

The giant Petrel *Macronectes giganteus* showed much higher Hg concentrations in the present study than the reports from the literature. In giant petrels from Bird Island, South Georgia, Hg concentrations in the liver were averaging 2.16 (males) and 4.95 (females) $\mu\text{g g}^{-1} \text{ dw}$ (González-Solís et al., 2002) i.e., from 5 to 10 times lower than our findings. Birds sampled in the South Shetland Islands and the Antarctic Peninsula had also Hg concentrations one order of magnitude lower, averaging $1.37 \mu\text{g g}^{-1} \text{ dw}$ (Szefer et al., 1993). This marked difference could be due to our small sampling number or to some local effect, since our samples present similar SIA levels to those from Bird Island, South Georgia, the closest site that could be found in the literature for comparison (Forero et al., 2005).

Hg concentrations in the eggs of Gentoos penguins *P. papua* and Adélie penguins *P. adeliae* were similar to that reported in the literature (Moreno et al., 1997, after dry weight conversion assuming 70% water content), i.e., $\sim 0.13 \mu\text{g g}^{-1} \text{ dw}$. This same work also presents data for *P. adeliae* liver, $\sim 1.5 \mu\text{g g}^{-1} \text{ dw}$, also in the same range with our findings. As for liver, literature shows more contaminated liver samples for two pygoscelids in Szefer et al. (1993), averaging 2.01 (*P. adeliae*) and $34.7 \mu\text{g g}^{-1} \text{ dw}$ (*P. papua*). However, Smichowski et al. (2006) presents data six times lower for *P. adeliae* chicks' liver collected also in King George Island, which provides evidence of Hg bioaccumulation in this species. Significant correlations for birds are broken down in Fig. 1. Only paired analyses are shown, therefore the number of samples may vary in regard to the total.

Since pygoscelids fast during the egg formation period (Astheimer and Grau, 1985; Polito et al., 2011), the influence on egg results of the trophic level of prey during breeding season is not to be considered. The significant negative correlation between Hg and $\delta^{13}\text{C}$

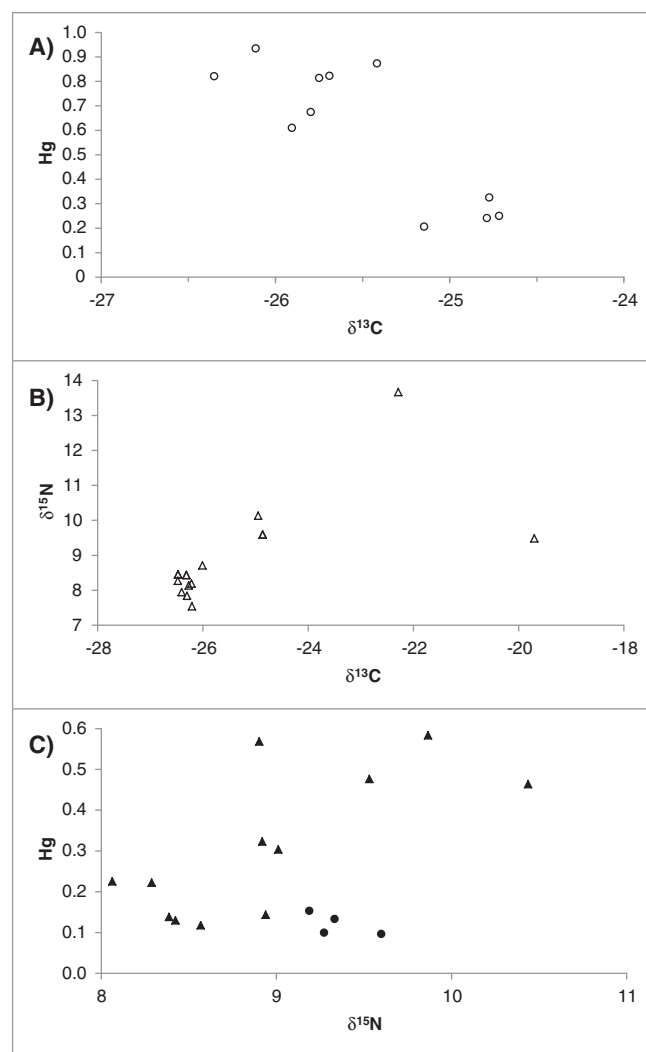


Fig. 1. Significant correlations between Hg ($\mu\text{g g}^{-1} \text{ dwt}$) and stable isotopes (‰) for A) *Pygoscelis antarctica* eggs; B) *P. antarctica* liver and C) *Pygoscelis papua* egg and liver. Empty markers for *P. antarctica*, filled ones for *P. papua*. Round markers for eggs, triangular ones for liver.

($\rho = -0.627$) in *P. antarctica* eggs could be related to the fact that fasting breeding adult penguins present a decrease in $\delta^{13}\text{C}$ plasma signature during fasting (Cherel et al., 2005), whereas Hg levels might increase simply due to the opposite of the growth dilution effect on egg-laying females and consequent transfer (e.g. Agusa et al., 2005). A significant negative correlation was found between Hg and $\delta^{15}\text{N}$ ($\rho = -0.900$) in *P. papua* egg could be related to different proportions of fish and squid in the diet prior to breeding (Polito et al., 2011). Since the Gentoos penguin presents considerable plasticity in its diet when compared to the other two less flexible pygoscelids (Miller et al., 2009), consuming prey from coastal/oceanic and benthic/pelagic origins could result in different $\delta^{15}\text{N}$ baselines and therefore misleading conclusions (see Chouvelon et al., 2012b). This is a similar issue as previously discussed for krill and limpets.

The significant positive correlation between $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ ($\rho = 0.562$) in *P. antarctica* liver is due to the concomitant enrichment of these elements even though it occurs in a much lesser degree for ^{13}C (DeNiro and Epstein, 1978). Finally, a significant positive correlation was found between Hg and $\delta^{15}\text{N}$ ($\rho = 0.615$) in *P. papua* liver likely due to the consumption of higher trophic level prey for adults all year round.

Hg concentrations for the eggs of the kelp gull *Larus dominicanus* fall within the same order of magnitude than the value (i.e., $0.46 \mu\text{g g}^{-1}$)

reported by Moreno et al. (1997) but data for deeper comparison are dramatically lacking for this species. However, Hg concentrations in the liver was the second highest value for seabirds, which is likely due to the influence of high consumption rates of *N. concinna* during the austral summer (Favero et al., 1997). The influence of this benthic prey can also be seen in the highly enriched $\delta^{13}\text{C}$ signature in kelp gull liver and moreover, in the resulting PCB congener profiles in *L. dominicanus*, already highlighted in a previous study (Cipro et al., 2013).

The Antarctic tern *Sterna vittata* is, in this study, a *sui generis* case, firstly because only one sample was available and secondly because its SIA results would suggest a coastal/benthic diet whereas the literature (Casaux et al., 2008) reports it as a pelagic predator. More samples are needed to explore this contradictory observation.

Pinnipeds will be further discussed as a group due to the very small and disperse sampling.

For animals interspecific data, two statistically (Tukey HSD tests) different intra-homogenous groups for Hg levels: one group with the giant petrel *Macronectes giganteus* liver and one group with all the other samples. This relative homogeneity around the Antarctic Peninsula had previously been reported by Brasso et al. (2012).

Particularly for pinnipeds, the data for *A. gazella* and *M. leonina* liver are clearly distinguished from the others. A previous analysis of the SIA data (Cipro et al., 2012) for *A. gazella* had already shown a clear shift in foraging area (closer to the shore) and towards higher trophic level, probably due to the increase of fish consumption. All of this corroborates this much increased Hg value, adding up to the fact that they stand on a higher trophic level and are more prone to biomagnification. Interpretation for *M. leonina* is yet more complicated since sex-specific foraging strategies are reported (Cherel et al., 2008; Lewis et al., 2006): in a general way females' diets vary less than the ones of males. Females forage mostly on low trophic level prey from deep waters, such as cephalopods or myctophid fish (according to the location), whereas, males appear to be specialist foragers, divided among several different strategies, which might also include pelagic invertebrates, benthic fish and adult pelagic fish. Such a difference can be seen as a strategy to avoid intra and interspecies competition, especially during the breeding season (Cherel et al., 2008; Lewis et al., 2006). Comparison with Szefer et al. (1993) shows that *Lobodon carcinophagus* muscle Hg concentrations are one order of magnitude lower and the ones of *Leptonychotes weddellii* kidney were two orders of magnitude lower. Since no sex, age or biometrics data could be taken, the interpretation cannot go any further.

Concerning intraspecific data for vegetation, Hg values for several moss species from Edmonson Point, Antarctica ranged from 0.05 to 0.15 $\mu\text{g g}^{-1}$ dw (Bargagli et al., 1998b), which comes in reasonable agreement with the data from the present study. Another work, in the same area as the present one (dos Santos et al., 2006) presents values for mosses of 0.023 (*Bryum* spp.) and 0.040 (*Polytrichum* spp.) $\mu\text{g g}^{-1}$, which are roughly 1.5 to 5 times lower than the ones hereby reported. Also, Hg concentrations of 0.036 $\mu\text{g g}^{-1}$ for lichens (*Usnea* spp.) are 4 times lower than the ones from the present work, in average. Significant positive correlation between Hg and $\delta^{13}\text{C}$ ($\rho = 0.828$) in *Sanionia uncinata* could be related to a lesser marine influence, as further discussed.

For vegetation, Tukey HSD tests separated the less Hg contaminated mosses *Syntrichia princeps* and *Sanionia uncinata* from the more Hg contaminated lichens *Usnea* spp. in two statistically different intra-homogenous groups. This finding comes in agreement with physiologic differences, mainly water dependence. An analogous separation between lichens and mosses is noticed in Cipro et al. (2011) for POPs. And since moister habitats and marine influence are related to lower $\delta^{13}\text{C}$ and higher $\delta^{15}\text{N}$ (Lee et al., 2009), it would be reasonable to assume that dry deposition phenomena are more significant as Hg source in this case. However, the clear influence of secondary sources in recent literature (Choy et al., 2010; Cipro et al., 2011; Roosens et al., 2007) might overcome that conclusion. Therefore our results differ largely from the

Table 2

Akaike information criterion for small sample sizes (AICc) on the two subsets of samples.

		AICc	ΔAICc	w_i
Animals				
$\delta^{15}\text{N}$	Tissue	20.93	0.00	0.59
$\delta^{13}\text{C}$	$\delta^{15}\text{N}$	22.84	1.91	0.23
$\delta^{15}\text{N}$	Species	25.27	4.33	0.07
$\delta^{15}\text{N}$	Species	25.35	4.41	0.06
$\delta^{13}\text{C}$	$\delta^{15}\text{N}$	27.07	6.14	0.03
$\delta^{13}\text{C}$	$\delta^{15}\text{N}$	27.18	6.25	0.03
Plants				
$\delta^{13}\text{C}$	Species	-72.82	0.00	0.68
$\delta^{13}\text{C}$	$\delta^{15}\text{N}$	-71.35	1.46	0.32
Species		-53.26	19.56	0.00
$\delta^{15}\text{N}$	Species	-51.19	21.62	0.00
$\delta^{13}\text{C}$		-29.53	43.29	0.00
$\delta^{15}\text{N}$		-28.60	44.22	0.00
$\delta^{13}\text{C}$	$\delta^{15}\text{N}$	-27.99	44.83	0.00

ones by dos Santos et al. (2006) not only in an intraspecific way, but also, and mainly, in the clear difference between lichens and mosses Hg levels.

In order to understand the influence of species, tissue, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ on how these variables affect the resulting Hg concentrations in this large array of samples, normal distribution and log-link function generalized linear models (GLM) were built as previously described. The whole dataset was split in animals and vegetation subsets and model results are shown in Table 2. Taking the results into account, the best model to explain Hg levels in the fauna is composed by both $\delta^{15}\text{N}$ and tissue, followed by $\delta^{13}\text{C}$, species and tissue. Examples of the importance of such variables in the accumulation of trace elements are found throughout the scientific literature: $\delta^{15}\text{N}$ (e.g. Anderson et al., 2010), tissue (e.g. Bustamante et al., 2003) and species (e.g. Borgå et al., 2006) as well as for both $\delta^{15}\text{N}$ and tissue (e.g. Campbell et al., 2005), as in our findings.

The models for vegetation, in turn, are composed by $\delta^{13}\text{C}$ and species followed by $\delta^{13}\text{C}$, $\delta^{15}\text{N}$ and species. Firstly, it is, in a broad sense, the repetition of Tukey HSD tests previously performed, since mosses and lichens were placed in two statistically different groups and moreover, a clear separation in $\delta^{13}\text{C}$ for lichens and mosses, with no overlapping whatsoever, can be noticed in Cipro et al. (2011). The influence of $\delta^{15}\text{N}$ in Hg concentrations, which in vegetation cannot be linked to trophic level is, firstly, linked to $\delta^{13}\text{C}$ (Cipro et al., 2011; Park et al., 2009) and also thought to be linked to the exposure to secondary contaminant sources, particularly the degradation of animal derived organic matter (Cipro et al., 2011).

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