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Baseline

Accumulation and potential sources of lead in marine organisms from coastal ecosystems of the Chilean Patagonia and Antarctic Peninsula area

Winfred Espejo[a,e], Janeide de A. Padilha[b], Rodrigo A. Gonçalves[c], Paulo R. Dorneles[b], Ricardo Barra[d], Douglas Oliveira[c], Olaf Malm[b], Gustavo Chiang[e], José E. Celis[f,*]

[a] Ecology & Biodiversity Department, Facultad de Ciencias de la Vida, Universidad Andrés Bello, Santiago, Chile
[b] Radioisotope Lab, Biophysics Institute, Federal University of Rio de Janeiro, Brazil
[c] Chemistry Department, Pontifical Catholic University of Rio de Janeiro, Brazil
[d] Department of Aquatic Systems, Facultad de Ciencias Ambientales y Centro EULA-Chile, Universidad de Concepción, Casilla 160-C, Concepción, Chile
[e] Melimoyu Ecosystem Research Institute, Lo Beltrán 2347 Vitacura, Santiago, Chile
[f] Department of Animal Science, Facultad de Ciencias Veterinarias, Universidad de Concepción, Casilla 537, Chillán, Chile.

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ABSTRACT

Environmental concentrations of Pb have been increased due to anthropogenic activities, which have provoked the released of this element to the environment in large amounts. To understand how Pb behaves in remote southern marine ecosystems, we measured Pb concentrations and isotope ratios in biota from coastal marine ecosystems of the Chilean Patagonia and the Antarctic Peninsula area. Lead concentrations in the aquatic organisms ranged from 0.02 to 1.19 mg kg^{-1} d.w. In Patagonia, higher Pb levels were found in fish than in invertebrates (crab, shrimp, porifera, limpet and shellfish). In comparison with the baseline reference values from other parts of the world, fishes exhibited lower Pb levels. The results of Pb isotopic compositions indicated that the bioaccumulation of Pb in marine organisms come from different anthropogenic sources. These Pb levels might be useful for further studies that allow a deeper evaluation of sources for Pb contamination in these remote ecosystems.

Coastal environments are very dynamic ecosystems, which are usually greatly impacted by humans. The quality of aquatic ecosystems is vital to ensure health protection to humans and wildlife. Lead is a toxic element that causes behavioral problems in vertebrates and can negatively interfere in growth rates, learning, and metabolism in general (Eisler, 1988; Weber and Dingel, 1997; Burger and Gochfeld, 2000). This metal is a well-known element whose environmental concentrations have been increased by anthropogenic activities worldwide. A variety of human sources of Pb to the environment include vehicular exhaust, leaded paint, and industrial emissions (Yu, 2001; Ahamed and Siddiqui, 2007).

It is quite difficult to identify the origins of Pb in the environment, thus it is extremely important to determine clearly which Pb emissions correspond to human activity or natural sources. Stable isotopes are a useful tool on tracking the origin of environmental contamination by Pb. Such use is based on the fact that the proportion of Pb stable isotopes is different between natural (e.g. erosion of rocky beds) and anthropogenic sources (Labonne et al., 1998; Labonne et al., 2001). Each Pb source has a different isotope ratio, which is very useful for identifying the origin of Pb in a sample (Gregurek et al., 1998; Komárek et al., 2008). Biological processes and physical-chemical fractionation do not significantly modify the proportions of Pb stable isotopes. The isotopes ^{206}Pb, ^{207}Pb and ^{208}Pb originate from the decays of ^{238}U, ^{235}U and ^{232}Th, respectively (Labonne et al., 1998). Some limitations can be addressed when using Pb stable isotopes, as a possible overlap of isotopic signatures or if the sources are not well defined. For that reason, it is suggested to use more than one kind of environmental sample (Sen et al., 2016). Pb isotopes have been successfully used on several studies that have dealt with different matrices (Hoven et al., 1999; Caurant et al., 2006; Philippe et al., 2008). Also, Pb stable isotopes have been used as fingerprints to study abnormal mortality of fauna after a storm water event by accumulation of this element (Raimundo et al., 2009).

Antarctica and Patagonia present marine ecosystems that have not been very affected by human presence, and they can be still considered to be low-anthropogenical-impacted regions. However, environments from both regions can be exposed to the impact of global anthropogenic activities (Commendatore and Esteves, 2007; Bargagli, 2008; Dorneles et al., 2015). The evidence indicates that global Pb sources can reach

* Corresponding author.
E-mail address: jcelis@udec.cl (J.E. Celis).

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remote areas through atmospheric transport from lower latitudes (Celis et al., 2015; Smichowski et al., 2006). Considering the increase of human activities in recent decades, Pb levels could increase in these areas. Benthic and pelagic organisms have been used to assess metal contamination in marine ecosystems elsewhere (Bargagli et al., 1998; Cheung et al., 2002; Deheyn et al., 2005; Ip et al., 2005; Gil et al., 2006; Dorneles et al., 2007; Comoglio et al., 2011; Conti et al., 2012; Primost et al., 2017). Stable Pb isotopes have been mostly used to study the sources of Pb in various abiotic samples (Wong et al., 2002; Liu et al., 2003; Wong et al., 2003: De Deckker et al., 2010; McConnell et al., 2014; Sen et al., 2016; Francová et al., 2017), but only a few studies have been done in biological samples to trace the anthropogenic origins of Pb elsewhere (Hoven et al., 1999; Ip et al., 2005; Raimundo et al., 2009; Primost et al., 2017) and less in remote areas (Outridge et al., 1997; Deheyn et al., 2005). Consequently, the objective of this study was to assess the accumulation and potential sources of Pb in macroinvertebrates and fishes from the coastal zone of Continental Chilean Patagonia and the Antarctic Peninsula area. These baseline results can be taken as reference values for further studies of environmental pollution.

During the austral summer 2013/2014, samples of macroinvertebrates and fishes were obtained from two sites of the western Chilean Patagonia (Sector 1, Fig. 1): Marchant River Mouth (44°5′S, 73°5′O) and Yalac Island (44°01′S, 73°14′W); both sites in Patagonia are characterized for being surrounded by abundant forests and native vegetation, far from industrialized areas, where there is only a small town of 57 inhabitants 4 km away. Also, biological samples were collected from two locations of the Antarctic Peninsula area (Sector 2, Fig. 1): Paradise Bay (64°51′S, 62°54′W; Gerlache Strait) and Cape Shirreff (62°28′S, 60°46′W; Livingston Island, South Shetland Islands); both Antarctic sites are isolated and distant from any human facilities. In Chilean Patagonia, 5 species of macroinvertebrates and 6 species of fishes were collected at Marchant River Mouth (n = 26), and 6 species of macroinvertebrates and 2 species of fishes were sampled at Yalac Island (n = 20; Table 1). In Antarctica, 6 species of macroinvertebrates and 3 species of fishes were sampled (n = 21) at Paradise Bay, and 4 species of macroinvertebrates (n = 6) at Cape Shirreff (Table 2).

Different species of fishes and invertebrates were collected from each location by scuba diving to ensure the collection of the selected species, as well as to minimize any impacts on the surroundings. Fishes were anaesthetized with BZ-20 (Veterquimica), sacrificed by severing the spinal cord, and sampled for muscle tissues. Soft tissues of mollusks were collected and whole bodies of other macroinvertebrates were retained. All specimens were stored at −20 °C until their processing in the laboratory.

All samples were freeze-dried until dry masses were constant and then were homogenized into a fine powder using a glass mortar and pestle pre-cleaned with 2% Conrad solution (Merck) for 24 h, washed with deionized water and HCl 1 M and then rinsed with distilled water (Wassenaar and Hendry, 2000). Sub-Samples (0.02 and 0.45 g) were digested in a microwave with high purity nitric acid (GR), hydrochloric acid and perchloric acid. The quality controls for the strong acid digestion method included reagent blanks and standard reference materials (NIST SRM 1566a and DORM-2). The recovery rates in the reference materials were around 80%.

The concentrations of Pb were determined by inductively coupled plasma mass spectrometry (ICP-MS, Elan 6000, Perkin Elmer-Sciex) at the Chemistry Department of the Pontifical Catholic University of Rio de Janeiro Rio de Janeiro, Brazil. An international standard reference material (NIST SRM 981, common lead) was used for calibration and

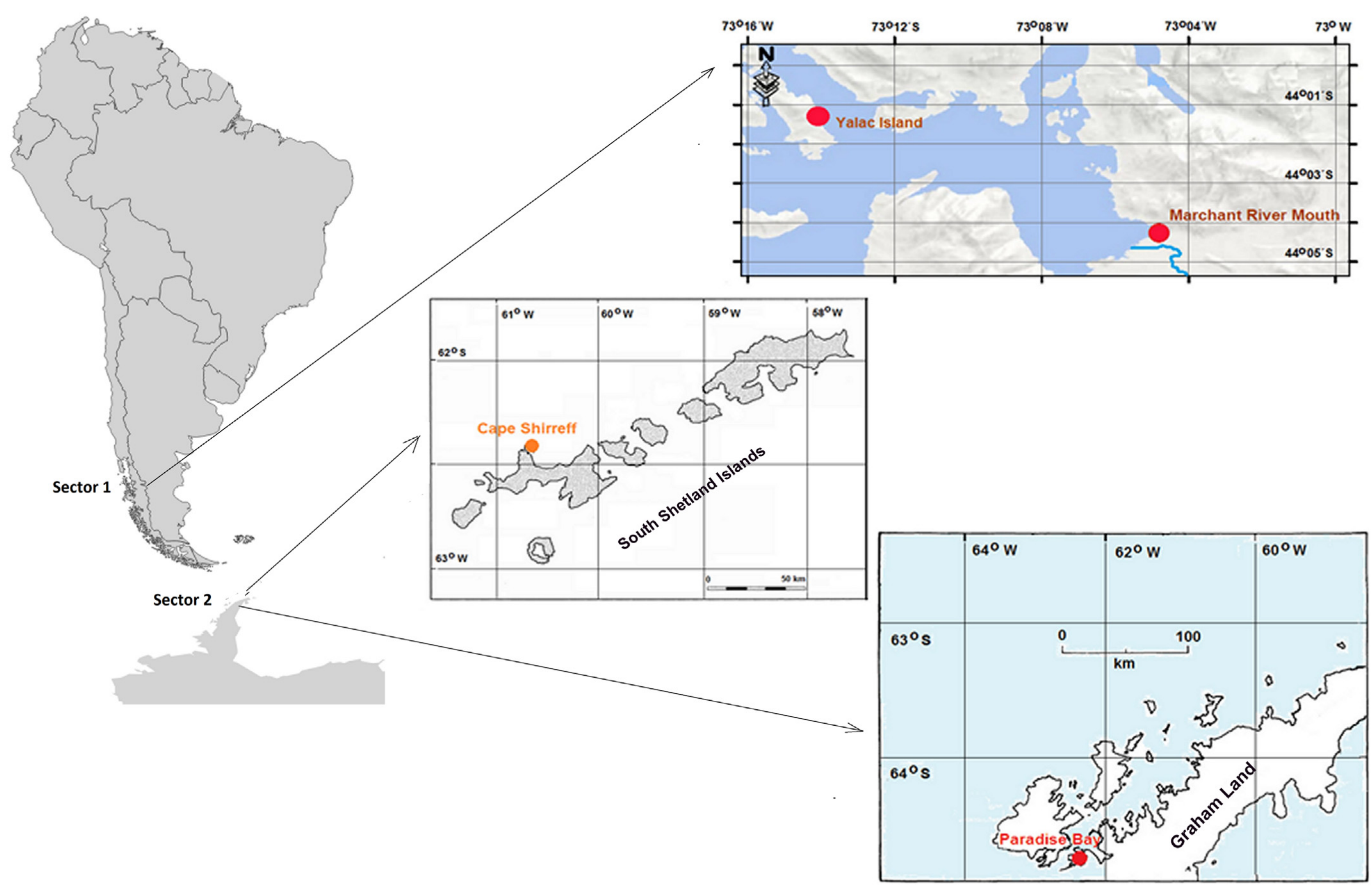

Fig. 1. Locations of the marine coastal ecosystems from the western Chilean Patagonia (Sector 1: Yalac Island and Marchant River Mouth) and Antarctic Peninsula area (Sector 2: Cape Shirreff and Paradise Bay).

Table 1
Pb concentrations (mg kg^{-1} d.w.) and ratios of $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$ in different groups of aquatic organisms from two locations of the western Chilean Patagonia coastline. Data are presented as mean ± standard deviation.

Location	Group	Species	*n*	Type of sample	Pb	$^{206}Pb/^{207}Pb$	$^{208}Pb/^{207}Pb$
Marchant River M.	Macroinvertebrates	*Stichaster striatus*	3	Soft tissue	0.18 ± 0.21	1.03 ± 0.02	1.01 ± 0.005
		Aulacomya ater	3	Soft tissue	0.20 ± 0.14	1.02 ± 0.01	1.00 ± 0.001
		Hemigrapsus granulosus	3	Whole body	0.15 ± 0.08	1.04 ± 0.03	1.02 ± 0.01
		Loxechinus albus	3	Soft tissue	0.06 ± 0.01	1.04 ± 0.02	1.03 ± 0.02
		Cancer coronatus	3	Soft tissue	0.47 ± 0.62	1.01 ± 0.02	1.01 ± 0.01
	Fishes	*Eleginops maclovinus*	3	Muscle	0.29 ± 0.39	1.12 ± 0.12	1.06 ± 0.06
		Genypterus blacodes	3	Muscle	0.04 ± 0.01	1.04 ± 0.03	1.01 ± 0.02
		Macruronus magellanicus	1	Muscle	1.00	1.03	1.00
		Merluccius australis	1	Muscle	1.01	1.03	1.01
		Salilota australis	1	Muscle	1.08	1.15	1.08
		Schroederichthys chilensis	2	Muscle	0.05 ± 0.05	1.05 ± 0.04	1.04 ± 0.05
Yalac Island	Macroinvertebrates	*Chorus giganteus*	3	Soft tissue	0.05 ± 0.03	1.08 ± 0.07	1.03 ± 0.02
		Concholepas concholepas	3	Soft tissue	0.02 ± 0.003	1.02 ± 0.09	1.02 ± 0.03
		Fissurella spp.	3	Soft tissue	0.02 ± 0.001	1.05 ± 0.04	1.00
		Nacella magellanica	3	Soft tissue	0.03 ± 0.01	1.08 ± 0.04	1.04 ± 0.02
		Cliona chilensis	1	Soft tissue	1.00 ± 0.22	1.01	1.00 ± 0.01
		Tegula atra	3	Soft tissue	0.39 ± 0.32	1.03 ± 0.01	1.02 ± 0.004
	Fishes	*Paralabrax humeralis*	1	Muscle	0.04	1.07	1.05
		Panguipes chilensis	3	Muscle	0.02 ± 0.003	1.08 ± 0.03	1.03 ± 0.02

Table 2
Pb concentrations (mg kg^{-1} d.w.) and ratios of $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$ in different groups of aquatic organisms from two locations of the Antarctic Peninsula area. Data are presented as mean ± standard deviation.

Location	Group	Species	*n*	Type of sample	Pb	$^{206}Pb/^{207}Pb$	$^{208}Pb/^{207}Pb$
Paradise Bay	Macroinvertebrates	*Diplasterias brucei*	2	Soft tissue	0.52 ± 0.59	1.04 ± 0.04	1.02 ± 0.02
		Chorismus antarcticus	1	Soft tissue	0.59	1.04	1.02
		Lyssianasid amphipod	3	Whole body	1.19 ± 0.69	1.03 ± 0.01	1.01 ± 0.003
		Nacella concinna	3	Soft tissue	1.10 ± 0.15	1.05 ± 0.003	1.02 ± 0.001
		Euphausia superba	3	Whole body	0.25 ± 0.23	1.03 ± 0.03	1.02 ± 0.02
		Haplocheira sp.	3	Whole body	0.22 ± 0.06	1.03 ± 0.01	1.02 ± 0.003
	Fishes	*Harpagifer antarcticus*	3	Muscle	0.15 ± 0.14	1.08 ± 0.01	1.03 ± 0.01
		Trematomus bernacchii	1	Muscle	0.03	1.06	1.06
		Trematomus hansoni	2	Muscle	0.03 ± 0.01	1.00 ± 0.001	0.99 ± 0.02
Cape Shirreff	Macroinvertebrates	*Diplasteria brucei*	1	Soft tissue	0.16	1.06	1.03
		Macroptychaster sp.	1	Soft tissue	0.09	1.03	1.01
		Nacella concinna	3	Soft tissue	0.49 ± 0.51	1.05 ± 0.02	1.03 ± 0.01
		Odontaster validus	1	Soft tissue	0.24	1.00	1.00

analytical control for the Pb isotopic measurements. All analyses were carried out in triplicate and the resulting values were averaged. Pb detection limit was 0.00011 mg kg^{-1}. Lead levels were expressed as mg kg^{-1} (dry weight), and then the ratios of $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$ were calculated.

The scarcity of scientific information on the Patagonian and Antarctic aquatic fauna in comparison with that of lower latitudes makes it difficult to discuss our results. In general, Pb concentrations ranged from 0.02 to 1.19 mg kg^{-1} (Table 1). The Pb concentrations verified in this study averaged 0.23 mg kg^{-1} in macroinvertebrates and 0.44 mg kg^{-1} in fishes from Patagonia. These data are lower than those presented by Ip et al. (2005) in macroinvertebrates and fishes from South China (0.16 to 122.8 mg kg^{-1} d.w., assuming a moisture content of 75% in the sample). The Pb levels in macroinvertebrates from Patagonia ranged from 0.02 to 1.0 mg kg^{-1}, being the lowest in the marine gastropod *Fissurella* sp. and the highest in the porifera *Cliona chilensis*, both found at Yalac Island. In Antarctica, the Pb levels in macroinvertebrates (0.09–1.10 mg kg^{-1}) were the highest in the sea star *Macroptychaster* sp. at Cape Shirreff, whereas the lowest were found in the limpet *Nacella concinna* at Paradise Bay. A recent study performed in Argentinean Patagonia revealed that Pb concentrations in gastropods varied from 1.02 to 10.37 mg kg^{-1} d.w. (Primost et al., 2017). Previously, Conti et al. (2012) found Pb levels ranging from 0.13 to 1.23 mg kg^{-1} d.w. in muscle and viscera of *Nacella magellanica* from Cape Horn (Southern Patagonia), while Comoglio et al. (2011) reported higher Pb levels (4.78 mg kg^{-1} d.w.) for the same species (whole body) and location. The Pb concentrations in fish muscle of the present study (0.03–0.15 mg kg^{-1}) were lower than those levels reported in the Antarctic Peninsula area (0.20–1.0 mg kg^{-1} d.w.) previously (Bargagli et al., 1998; Deheyn et al., 2005; Beltcheva et al., 2011). The Pb levels measured in mollusks are lower than those levels allowed for human consumption (1.5 mg kg^{-1}) according to European regulations (Gil et al., 2006) and those recommended by the Joint FAO/WHO Expert Committee on Food Additives & World Health Organization (2006). Those regulations are based on soft tissues of mollusks. However, it is necessary to state that some species of macroinvertebrates are not consumed whole by humans. In fish, the highest Pb concentration was found in *Salilota australis* (1.08 mg kg^{-1}), and was below the permissible limits in fish proposed by European Commission (2006).

In the Chilean Patagonia, the $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$ ratios of the marine organisms ranged from 1.01 to 1.15 and 1 to 1.08, respectively (Table 1). The aquatic organisms from Marchant River Mouth exhibited wider $^{206}Pb/^{207}Pb$ (1.01–1.15) and $^{208}Pb/^{207}$ ratios (1.0–1.08) than those from Yalac Island. In the Antarctic Peninsula area (Table 2), the $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$ ratios of the marine organisms ranged from 1.0 to 1.08 and 0.99 to 1.06, respectively. The marine organisms from Paradise Bay showed wider $^{206}Pb/^{207}Pb$ (1.0–1.08) and $^{208}Pb/^{207}$ ratios (0.99–1.06) than those from Cape Shirreff. These ratios are lower than $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$ ratios of similar aquatic organisms reported from South China, which ranged from 1.16 to 1.19 and 2.44 to 2.49, respectively (Ip et al., 2005).

The Pb accumulated in aquatic organisms can result from natural

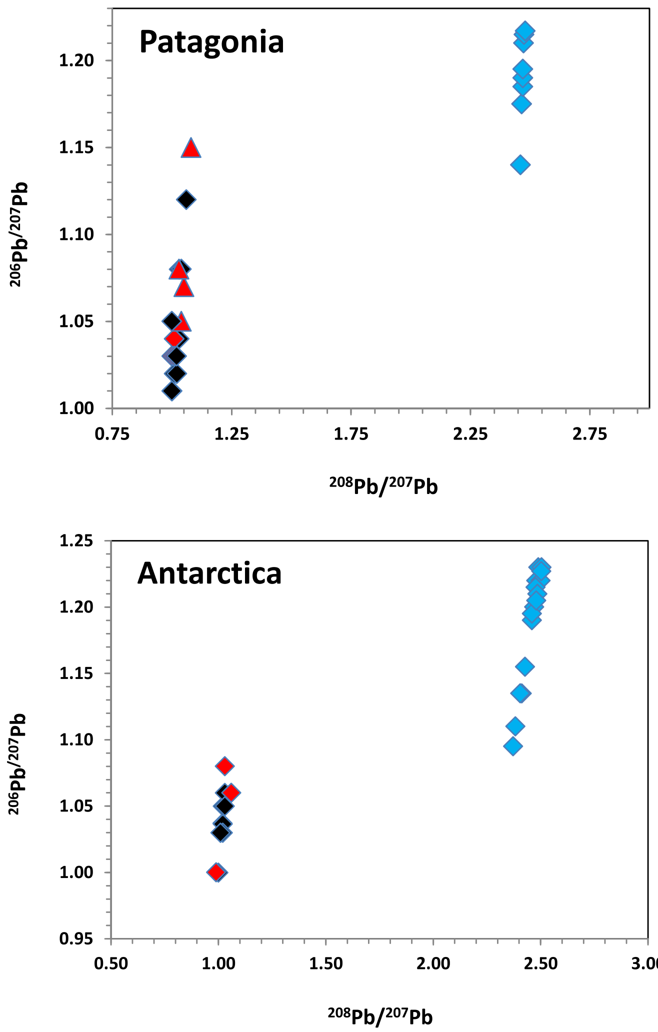

Fig. 2. Plot of the $^{208}Pb/^{207}Pb$ versus $^{206}Pb/^{207}Pb$ averaged values obtained from macroinvertebrates (black dots), fishes (red dots) of the Patagonia and Antarctic Peninsula area. Background geological materials (blue dots) are values reported by De Deckker et al. (2010). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

sources (weathering, erosion, and transport from bedrocks) and from anthropogenic activities [e.g. atmospheric or advective transport (through water)]. The background Pb isotopic ratios of abiotic materials from Antarctica ranged from 1.10 to 1.23 for $^{206}Pb/^{207}Pb$, whereas for $^{208}Pb/^{207}Pb$ ratio the range varied between 2.37 and 2.52 (Planchon et al., 2003; De Deckker et al., 2010; McConnell et al., 2014). In Patagonia, the $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$ ratios of geological materials ranged 1.14–1.22 and 2.46–2.48, respectively (De Deckker et al., 2010). As shown in Fig. 2, all marine organisms from Patagonia and Antarctica exhibited lower Pb isotopic values than those found in geological material and air deposition, thus suggesting that the sources of lead for the marine biota of those two regions were mainly of anthropogenic origin. The presence of Pb in remote areas of the Southern Hemisphere can be due to a more activated dust pathway, as it is now clear that some air masses from Australia can reach Patagonia and even Antarctica (De Deckker et al., 2010). There is evidence indicating that anthropogenic Pb is well mixed in the Antarctic atmosphere, thus deposition of this toxic metal occurs across the continent (McConnell et al., 2014).

According to our knowledge, this is one of the first reports of lead isotope ratios in marine species from the Chilean Patagonia and the Antarctic Peninsula area. There are still scarce studies of lead isotopes in aquatic organisms. However, the Pb isotopic signature has been shown to be an important tool for environmental studies (Caurant et al., 2006; Philippe et al., 2008; Raimundo et al., 2009). Data on Pb isotopic signature can be useful for different disciplines. They can be useful in geobiology in order to study the interaction of the aquatic organisms with the physical-chemical characteristics of the environment. In biological studies they can be used to identify differences between species or to envision possible changes in their composition. Regarding the data generated by the present study specifically, they can be used in future environmental science studies, for comparing Pb isotopic signatures between remote areas and regions under extreme anthropogenic pressure.

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