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Concentrations of lead in pinniped bones confirm Galapagos as a relatively unpolluted environment

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ABSTRACT

Lead (Pb) is a trace element that is naturally present in arid regions but it is also released to the marine environment by anthropogenic industrial emissions. Here, we assessed Pb concentrations in bone samples of four pinniped species: the Galapagos sea lion *Zalophus wollebaeki*, sampled in Galapagos archipelago, the monk seal *Monachus monachus* from Mauritania, and the South American fur seal *Arctocephalus australis* and the South American sea lion *Otaria flavescens*, from Uruguay, and investigate potential geographic differences. Concentrations of lead in the samples from Galapagos were lower than those detected in samples from Mauritania and Uruguay, indicating that the Galapagos archipelago is a comparatively pristine spot for this toxic element as related to the other two areas. The waters of Mauritania and Uruguay are likely affected by the inputs of lead brought by the desert dust and released by the local industry, respectively. This study supports the use of bone to assess lead concentrations in biota, as well as the use of pinnipeds as bioindicators of marine pollution.

1. Introduction

Lead (Pb) is naturally present in the environment in the chemical forms of Pb salts, ionic Pb, tetraethyl Pb, or Pb bound to other organic molecules (Assi et al., 2016). Pb may also originate from anthropogenic sources, mainly from the steel industry and mining, among other industrial activities (Rodríguez and Mandalunis, 2018), and is used in diverse applications, such as in the manufacture of batteries, electrical systems, construction materials and, in the past, as an additive to gasoline (Avery and Watson, 2009; Bridgestock et al., 2016, 2018; Järup, 2003; Pacyna and Pacyna, 2001). During the 20th century, the Pb input into the oceans caused by anthropogenic activities far exceeded natural concentrations (Boyle et al., 2014).

In vertebrates, Pb is deposited mainly in the bone matrix due to its ability to replace cations of Ca^{2+}, Mg^{2+}, Fe^{2+} and Na^{+} (Rodríguez and Mandalunis, 2018). Lead pollution has been linked to detrimental effects on the health of wildlife, the environment, and public health. In humans, elevated exposure to this metal has been associated to hypertension, anaemia, risk for stroke, neurotoxicity, hypocalcemia and hypophosphatemia (Dongre et al., 2013; Gambelunghe et al., 2016; Vorvolakos et al., 2016). Laboratory tests on rodents determined that the threshold value associated with adverse effects of Pb on bone formation is 50 mg kg^{-1} dw (Andrews et al., 1989; Carmouche et al., 2005; Lanocha et al., 2012).

The identification of heavy metal pollution hot spots and potential hazards to human health is usually carried out through the monitoring of concentrations in biota. In the case of marine ecosystems, the most common procedure is to analyse bioindicators such as mussels or other shellfish. However, because their distribution is limited to the continental shelf, their coverage is restricted to inshore coastal waters (Borrell et al., 2014). Analogous information on oceanic waters is much scarcer, as adequate indicators are not commonly available. Within this context, mobile (but local) top predators such as marine mammals have been proposed as potential indicators for trace metals exposure (*e.g.*, Vighi et al., 2017; De María et al., 2021; Borrell et al., 2023).

Pinnipeds are potential bioindicators of marine pollution, since they

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have been successfully used for other groups of environmental pollutants (*e.g.*, Garcia-Garin et al., 2020; Perez-Venegas et al., 2020). Although bone has been scarcely used as a target tissue for cetacean and pinniped research (Borrell et al., 2023; De María et al., 2021; Garcia-Garin et al., 2021; Hao et al., 2020; Honda et al., 1986; Lavery et al., 2008; Szteren and Aurioles-Gamboa, 2013; Vighi et al., 2017), it is a promising tissue to analyse Pb levels because, as stated above it accumulates mainly in the bone. Therefore, the Pb concentration in the bones of pinnipeds living in marine areas subject to different degrees of anthropogenic pressures would likely indicate differences in environmental Pb concentrations.

In the present study, the bone of four pinniped species, namely: the Galapagos sea lion *Zalophus wollebaeki*, the monk seal *Monachus monachus*, the South American fur seal *Arctocephalus australis* and the South American sea lion *Otaria flavescens*, were analysed to assess the concentration of Pb in three marine areas: (1) The Galapagos archipelago (Ecuador), a UNESCO World Heritage Site well-known for its endemic biodiversity and where the only anthropogenic impact is related with tourism; (2) the waters off Mauritania, in the Eastern coast of North Atlantic Ocean, where dominant winds transport the dust from the Sahara desert to the sea; and (3) the Uruguay coast in the Western coast of South Atlantic Ocean, which receive the waters from the Rio de la Plata estuary affected by anthropogenic activities producing Pb inputs, such as industrial outfalls and mining. The Rio de la Plata is one of the largest estuarine systems of South America and is the fifth largest in the world and drains the second largest basin in South America.

2. Material and methods

2.1. Study area and sampling

In the Galapagos archipelago, the bone samples (a small fragment of turbinate bone taken from the nasal cavity) were collected from 20 individuals of Galapagos sea lion (10 males and 10 females) found dead between 2000 and 2001 at the breeding sites of five islands (see Drago et al., 2016 and Fig. 1). In Mauritania, samples were collected from 11 individuals of monk seal (unknown sex) stranded dead in 1997 along the coastline (Fig. 1; See Pinela et al., 2010). In Uruguay, samples were collected between 2006 and 2008 from 24 individuals of two species, South American fur seal (n = 12; 6 males and 6 females) and South American sea lion (n = 12; 6 males and 6 females), found stranded dead along the coastline (Fig. 1), incidentally caught by fishermen, or obtained from skulls preserved in the scientific collection of the *Museo Nacional de Historia Natural* and the *Universidad de la República* at Montevideo, Uruguay (see Drago et al., 2020). The exact age of the sampled individual was unknown to us, although we only analysed skulls from physically mature specimens in order to minimize any potential age-related bias. The four populations considered in the present study are non-migratory animals (Drago et al., 2016; González Carman et al., 2016; Pinela et al., 2010; Riet-Sapriza et al., 2013; Rodriguez et al., 2013). Therefore, the Pb concentrations found in their bones can serve as potential indicators of the amount of Pb present in their living areas.

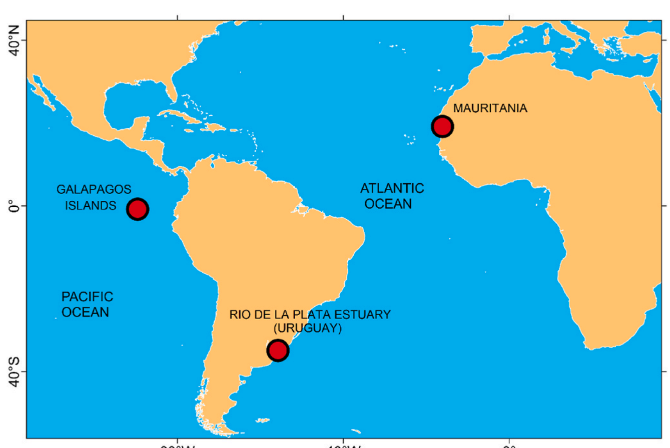

Fig. 1. Study area and sampling locations shown by red dots. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

2.2. Lead analysis

Lead analysis was performed following Garcia-Garin et al. (2021): 0.1 g of each powdered bone sample was acid-digested in clean Teflon reactors using 2 mL of HNO_3 (70%) and 1 mL of H_2O_2 (30%). After 12 h incubation at 90 °C, digested samples were diluted in 46 mL distilled water. Subsamples (10 mL) of each diluted sample were analysed with an ICP-MS (Induction Coupled Plasma-Mass Spectrometer) Nexion 350 PerkinElmer. One blank and the Bone Meal 1486 standard reference material, as certified by the US National Institute of Standards and Technology (NIST), were analysed every 10 samples to validate analyses. Lead concentrations were expressed as mg kg^{-1} dry weight (dw). The recovery percentage ranged from 99 to 103%. The lowest -Limit of Quantification- (*i.e.*, LOQ) was 0.05 mg kg^{-1} dw. Analyses were performed at the *Centres Científics i Tecnològics* (CCiT-UB) of the University of Barcelona, Spain.

2.3. Statistical analysis

The normality and heteroscedasticity of the distribution of Pb concentrations were preliminary tested using the Shapiro Wilk and Levene's tests, respectively. As the tests showed that data distribution departed from normality, comparisons were made using the non-parametric Kruskall-Wallis rank sum test and the post-hoc Dunn test, using the Benjamini-Hochberg method. The significance level was set at p-value <0.05. Calculations were carried out with the programming environment R (R Core Team, 2022).

3. Results and discussion

Lead was detected in all samples at concentrations ranging between 0.05 and 175.14 mg kg^{-1} dw (Table 1). This range was consistent with concentrations previously found in other pinniped studies conducted in calcified matrices (Table 1). The Pb concentrations here detected do not appear to be sufficiently high to produce adverse effects (Andrews et al., 1989; Lanocha et al., 2012), except for one South American sea lion whose bone Pb concentration reached 175.14 mg kg^{-1} dw. This individual presented Pb pellets lodged in its skull, an evidence of human induced-trauma likely due to conflictive interactions with recreational fisheries, although after forensic analysis, it was not safe to assert that the individual died from that cause. This concentration far exceeded the generally accepted 50 mg kg^{-1} dw toxicity thresholds in rodents (Andrews et al., 1989; Lanocha et al., 2012), and for this reason it is believed likely that it may have caused effects to the individual.

Our results showed that the lead concentrations in the bone of pinnipeds from the Galapagos Islands were lower than those in the bones of individuals from Mauritania and the Rio de la Plata estuary (p-value <0.0001; Table 1; Fig. 2). This is clearly justified by the remoteness and isolation of the Galapagos archipelago in relation to the other geographic areas. Also, the local management practices, environmental control and moderate population density of the archipelago all contribute to limit the sources of local pollution (Alava et al., 2014; Alava and Ross, 2018) although it is also true that other types of pollutants (*e.g.*, marine litter, PCBs, DDTs, and PBDEs) have been reported to be in the rise in the last decades as a consequence of the increase in tourism and human population (Jones et al., 2021; Alava et al., 2022). The remoteness and low industrialization would also apply to

Table 1
Number of samples and lead concentrations (mean ± SD, median, max and min values, expressed in mg/kg dw) detected in the bone of pinniped species from Galapagos archipelago, Mauritania and Uruguay, and those currently available in the literature in other pinniped studies conducted in calcified matrices. [a]indicates wet weight.[b] indicates mean ranges.

Species	Area	n	Tissue	Mean ± SD	Median	Min.	Max.	Reference
Zalophus wollebaeki	Galapagos archipelago	20	Bone	0.12 ± 0.07	0.10	0.05	0.37	This study
Monachus monachus	Mauritania	11	Bone	1.80 ± 1.93	0.90	0.54	5.92	This study
Arctocephalus australis	Rio de la Plata estuary	12	Bone	2.92 ± 4.35	1.70	0.27	16.27	This study
Arctocephalus australis	Rio de la Plata estuary	61	Teeth	0.17 ± 0.12	–	–	–	De María et al. (2021)
Otaria flavescens	Rio de la Plata estuary	12	Bone	18.74 ± 49.95	1.14	0.44	175.14	This study
Otaria flavescens	Rio de la Plata estuary	33	Teeth	0.42 ± 0.41	–	–	–	De María et al. (2021)
Zalophus californianus	California	6	Bone	8.7–34.2[b]	–	–	–	Braham (1973)
Zalophus californianus	California	6	Teeth	12.9[b]	–	–	–	Braham (1973)
Leptonychotes weddellii	Antarctica	2	Bone	0.07 ± 0.11[a]	0.07	0.03	0.48	Yamamoto et al. (1987)
Arctocephalus gazella	Bird Island	4	Teeth	20–150[b]	–	–	–	Cruwys et al. (1994)
Phoca hispida	North Atlantic	16	Teeth	40–60[b]	–	–	–	Cruwys et al. (1994)
Odobenus rosmarus rosmarus	North Atlantic	7	Teeth	8.2 ± 7.7	–	1.7	17.3	Outridge et al. (1997)
Odobenus rosmarus rosmarus	North Atlantic	79	Teeth	0.89–2.31[b]	–	–	–	Outridge and Stewart (1999)
Callorhinus ursinus	Northern Japan and Okhotsk Sea	16	Teeth	–	–	–	–	Arai et al. (1999)
Eumetopias jubatus	North Pacific	63	Teeth	10.04 ± 11.6	–	2.08	39.50	Ando et al. (2005)
Odobenus rosmarus divergens	North Pacific	199	Teeth	1.15 ± 0.54		0.29	4.60	Jay et al. (2008)
Phoca vitulina	North Sea	58	Bone	0.11	–	–	–	Agusa et al. (2011)

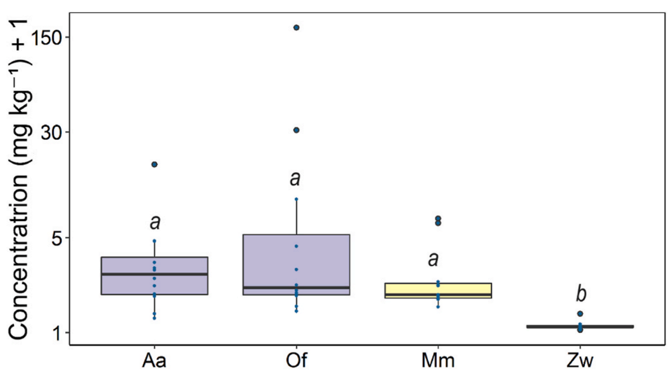

Fig. 2. Box-plots of lead concentrations detected in the bone of the four pinniped species analysed. In violet the South American fur seal (Aa) and South American sea lion (Of) from Rio the la Plata estuary (Uruguay), in yellow the monk seal (Mm) from Mauritania and in black the Galapagos sea lion (Zw) from the Galapagos archipelago (Ecuador). Horizontal lines represent medians, boxes represent interquartile intervals, and whiskers represent values within 1.5 times the interquartile range from the boxes. Outliers are plotted as larger points. The "y" axis is shown in natural logarithmic scale. Boxplots indicated by different lowercase letters are statistically different in their median values according to Kruskal-Wallis and Dunn's tests. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Mauritanian waters, but there the geographical and ecological characteristics are quite different. The Sahara dust, carried by the wind, is a significant source of ocean contamination, particularly of metals such as Pb (Abiye et al., 2020; Garrison et al., 2006; Morman et al., 2013), and this is clearly reflected in the bone of the local *M. monachus* population. The Pb concentrations found in the two pinniped species sampled in the Rio de la Plata estuary were similar to those detected in the species from Mauritania, although higher than those of the Galapagos archipelago but there the source of contamination is very likely different. As the contribution of wind-blown dust does not appear to be relevant along that segment of the coast, the high concentrations observed in South American sea lions and fur seals could be of anthropogenic origin and derive from the intense industrial activity present in the area (García-Rodríguez et al., 2010). The contaminants produced by the industries, including Pb, are transported by rivers and the atmosphere to the Rio the la Plata estuary and, as both species are known to use estuarine habitats and feed on coastal fish (Naya et al., 2002; Drago et al., 2020), the exposure to the local sources of pollution are facilitated. Further studies are necessary to trace the anthropogenic origins of Pb in samples.

4. Conclusions

This study shows that the use of pinniped or other marine mammal species as bioindicators of marine pollution is a practical approach to assess Pb concentrations in separate marine areas, as has proven useful previously in the monitoring of other contaminants (Aguilar and Borrell, 2005; Vighi et al., 2017). It also shows that the Galapagos archipelago is a pristine environment for Pb pollution as compared with the coasts of Mauritania and the Rio de la Plata estuary, which are highly impacted by natural and anthropogenic sources of Pb, respectively. The present study supports the validity of bone to assess lead concentrations in marine organisms and, by extension, in their environment. Additional research is required to elucidate the anthropogenic origin of Pb in the samples. Analysing Pb within the trophic web of these areas could contribute to clarifying these differences.

CRediT authorship contribution statement

Odei Garcia-Garin: Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Asunción Borrell:** Writing – review & editing, Supervision, Resources, Conceptualization. **Alex Aguilar:** Writing – review & editing, Resources. **Morgana Vighi:** Writing – review & editing, Supervision. **Meica Valdivia:** Writing – review & editing, Resources. **Enrique M. González:** Writing – review & editing, Resources. **Diego Páez-Rosas:** Writing – review & editing, Resources. **Massimiliano Drago:** Writing – review & editing, Resources, Project administration, Funding acquisition, Conceptualization.

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.

Data availability

Data will be made available on request.

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