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Variation of 210-polonium in the cephalopod community from the Bay of Biscay, North-East Atlantic

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Abstract: Among natural radionuclides, ^{210}Po is the major contributor to the radiation dose received by marine organisms. In cephalopods, ^{210}Po is concentrated in the digestive gland, which contains over 90% of the whole-body burden of the nuclide. Although previous studies showed that ^{210}Po was taken up independently of ^{210}Pb , its parent nuclide, very little is known about the factors influencing its levels in cephalopods. To the best of our knowledge, no studies investigated ^{210}Po levels in different species at the same time. In the present study, ^{210}Po was analysed in the digestive gland of 62 individuals from 11 species representing a large range of feeding ecologies and habitats, including squids, cuttlefish and octopus species from coastal to deep-oceanic habitats. Among species, the highest activity was measured in *Loligo vulgaris* (5720 ± 3606 Bq/kg) and the lowest in *T. megalops* (188 Bq/kg). However, considering the habitats (benthic vs pelagic and neritic vs oceanic), no significant differences appeared. At the species level, no differences between sexes were found so both sexes were plotted together to test the size effect for species with at least 8 individuals (i.e., *Eledone cirrhosa*, *L. vulgaris*, *L. forbesi* and *Sepia officinalis*). In the first three species, ^{210}Po levels decreased significantly with increasing size or weight but not in *S. officinalis*. In squid, this could be related to ontogenetic changes in diet from a high proportion of crustaceans (high Po content) in small individuals to fish (low Po content) in larger individuals, while the high dietary plasticity of *S. officinalis* at all stages of its life cycle could explain the lack of decrease in ^{210}Po with size. In comparison to the few data from the literature, the levels of ^{210}Po concentrations in the cephalopod community of the Bay of Biscay were overall in the same range than those reported in other cephalopods, varying across 4 orders of magnitude. Further studies are needed to understand the mechanism of retention in the cephalopod digestive gland.

Keywords: Bioaccumulation; Polonium; Squid; Octopus; Cuttlefish; trophic transfer

1. Introduction

Cephalopods have the ability to concentrate contaminants in their tissues to very high concentrations (e.g., [Penicaud et al. 2017](#); [Rodrigo and Costa 2017](#)). In this respect, the digestive gland appears to be a key organ in the metabolism of organic, metallic and radioactive contaminants (e.g., [Martin and Flegal, 1975](#); [Ueda et al. 1979](#); [Guary and Fowler 1982](#); [Bustamante et al. 2002a](#); [Ueno et al. 2003](#)). Thus, this organ is deeply involved in the processes of assimilation, detoxification and retention of contaminants, being the storage tissue for organochlorinated compounds and trace elements (e.g., [Tanabe et al. 1984](#); [Miramand and Bentley, 1992](#); [Bustamante et al. 2002b](#); [Danis et al. 2005](#)). The digestive gland of cephalopods also concentrates natural radionuclides such as polonium 210 (^{210}Po) to very high levels compare to the other tissues ([Heyraud and Chery 1979](#); [Smith et al. 1984](#); [Waska et al. 2008](#)). This organ contains over 90% of the whole-body burden of the nuclide in cephalopods, which is the most dominant radionuclide within the marine tropic pathway that results in ^{210}Po accumulation in marine organisms ([Carvalho and Fowler, 1993](#)). Previous studies showed that ^{210}Po was taken up independently of ^{210}Pb , its parent nuclide ([Smith et al. 1984](#); [Fowler 2011](#)), but very little is known about the factors influencing ^{210}Po levels in cephalopods. In planktonic crustaceans as well as in fish, size appears a key factor determining ^{210}Po levels in the organisms ([Cherry and Heyraud 1988, 1991](#); [Strady et al. 2015](#)). In cephalopods, the relationships with size do not appear so clear, with negative and positive relationships considering muscle or whole organisms, respectively ([Heyraud et al. 1994](#)). More broadly, little information is available concerning ^{210}Po in cephalopods as investigations to date mainly focused on a limited number of commercial species such as the common cuttlefish *Sepia officinalis* and the European squid *Loligo vulgaris* (e.g., [Heyraud and Cherry 1979](#); [Smith et al. 1984](#); [Waska et al. 2008](#)). Cephalopods are a class of marine molluscs that are widespread in diverse marine habitats from polar to tropical environments and from coastal waters to the deep-sea ([Boyle and Rodhouse,](#)

2005). Thus, they occupy benthic (e.g. octopus), nectobenthic (cuttlefish, bottlesquid), neritic and oceanic (mainly squids) habitats from the surface to deep waters and have a large variety of trophic niches. Importantly, they constitute a primary food source for many marine predators such as fish, marine mammals or seabirds, for instance the swordfish *Xiphias gladius*, the long-finned pilot whale *Globicephala melas* or the wandering albatross *Diomedea exulans* (Clarke 1996; Croxall & Prince 1996; Klages 1996; Smale 1996). Due to their key position in marine food webs, they represent a major vector of contaminants to their predators, as previously shown for cadmium (Bustamante et al. 1998). However, estimation of marine predator exposure to ^{210}Po following predation on cephalopods is limited due to paucity of knowledge regarding their tissue-burden levels and bioavailability as prey for higher trophic organisms. In humans, exposure to ^{210}Po through the consumption of cephalopods is generally limited, given that polonium concentrations are relatively low in the edible parts (i.e. muscles) compared with other edible seafood (e.g. oysters, mussels, scallops, all of whose soft tissues are consumed; Belivermiş et al., 2019, Bustamante et al. 2002c). Similarly, only the flesh of fish is consumed contributing to a low polonium intake for human consumers (Kılıç et al., 2018). However, unlike fish gut, the digestive gland of cephalopods is consumed in some countries such as in Japan where it is used for making a widely consumed dish named shiokara or consumed directly from very fresh squids. Thus, the consumption of cephalopod digestive gland may also be of direct concern to public health and more knowledge and research into the contents and potential risks to human consumers from consuming ^{210}Po from cephalopods is required.

Among natural radionuclides, ^{210}Po is the major contributor to the radiation dose received by marine organisms. In addition to its high specific activity and assimilation efficiency (Fowler 2011; Belivermiş et al., 2019), ^{210}Po is a high energy alpha particle emitter (5.3 MeV; Cherry and Shannon 1974) that can cause intense ionization of tissues. It can thus account for until

80% of the committed efficient dose due to natural radionuclides in human consumers of seafood (Piñero-García et al., 2022).

In this context, the present study aims at providing baseline data on ^{210}Po levels in cephalopods for a large range of species from the North-eastern Atlantic waters and at investigating the factors (e.g., sex, size, taxonomy) influencing their levels. To this end, ^{210}Po levels have been determined in the digestive gland of several cephalopod species, including squid, cuttlefish, and octopus caught in the Bay of Biscay, including coastal to deep-waters species.

2. Materials and Methods

2.1. Study area

Fieldwork was carried in the Bay of Biscay (from 1 to 10° W and from 43 to 48° N) opening onto the North-East Atlantic Ocean between France and Spain (Fig. 1). The Bay of Biscay is a very large bay of more than 220 000 km² with a continental shelf extending more than 200 km offshore in the north of the Bay but only 10 km in the south. Two main river plumes, i.e. the Loire and the Gironde, influence its hydrological structure (Planque et al., 2004; Puillat et al., 2004). The Bay of Biscay is home to a rich fauna including many protected species, such as sharks, rays, seabirds and marine mammals (Lassalle et al. 2014). It is subject to numerous anthropogenic activities such as extensive fishery activity (Lorance et al., 2009; OSPAR, 2010).

2.2. Sampling and sample preparation

Sampling was carried out in October-November 2011 during the EVHOE (“EVALuation Halieutique de l’Ouest de l’Europe”; <https://doi.org/10.17600/11040060>) groundfish survey conducted by Ifremer (“Institut Français de Recherche pour l’Exploitation de la Mer”). During this survey, bottom and pelagic trawls were performed on the continental shelf, on the slope

and in the canyons indenting the continental slope to specifically collect neritic, oceanic and deep-sea organisms.

For the present study, selected cephalopods included 62 individuals belonging to 11 species (Table 1). Cephalopod species were caught in various habitats (inshore vs. offshore waters, pelagic vs. benthic or demersal domains). They belong to six squid species: the veined squid *Loligo forbesi*, the European squid *L. vulgaris*, the European flying squid *Todarodes sagittatus*, the lesser flying squid *Todaropsis eblanae*, the Atlantic cranch squid *Teuthowenia megalops*, the reverse jewel squid *Histioteuthis reversa*; one bobtail species: the stout bobtail *Rossia macrosoma*; one cuttlefish species: the common cuttlefish *Sepia officinalis*; and three octopus species: the common octopus *Octopus vulgaris*, the spider octopus *Octopus salutii*, the horned octopus *Eledone cirrhosa*.

Each individual was frozen at -20°C on board and back to LIENSs laboratory. They were then thawed and dissected to remove the digestive gland. Wet weight (g), dorsal mantle length (mm) for all specimens and total length (for octopuses, in mm) were determined as well as the sex by direct observation of the gonads.

Following dissection, digestive gland samples were frozen at -20°C in individual plastic bags and freeze-dried for 48h. Freeze-dried samples were ground into a fine powder with porcelain mortar and pestle. The homogenised samples were then stored individually in plastic vials.

2.2. Radionuclide determination

Subsamples of ~ 1 g dry weight (dw) of cephalopod digestive gland were spiked with ^{209}Po yield tracer (ca. 50 mBq; $T_{1/2} = 102$ years) to assess the chemical yield and calculate the activity concentration of ^{210}Po . The internal standard was prepared with a certified standard solution of ^{209}Po (Eckert & Ziegler® Isotope Products). They were then digested adding 2 mL of 65% HNO_3 and 2 mL of 30% H_2O_2 in round bottom flasks in a hot plate with temperature control

(80°C). The solution was evaporated near to dryness to remove the acids, a step which was repeated 3 more times. The residues were then dissolved in 5 mL 0.5M HCl. Ascorbic acid was added until the yellow colour faded permanently, and polonium was allowed to deposit spontaneously onto silver discs for 8 h (Flynn, 1968). Measurements were performed with a high-resolution alpha spectrometer (CANBERRA) with passivated implanted planar silicon (PIPS) detectors with an active area of 450 mm² and 17.6 keV nominal resolution. Samples were measured until at least 400 counts were observed for individual ²¹⁰Po peak. The characteristic peak areas of the alpha particles from ²¹⁰Po and the tracer ²⁰⁹Po activity at the energies of 5.30 MeV and 4.88 MeV were used in the analysis of the sample spectra. The Certified Reference Material (CRM) IAEA-437 (Mussel from Mediterranean Sea) was used for accuracy control. CRM samples were processed following the same way as the cephalopod samples. The recovery obtained for ²¹⁰Po in the CRM was 90 ± 3%.

2.3. Statistical analysis

Descriptive statistics were applied using Minitab 13.1. Differences between species, habitats, and between males and females were determined using Mann-Whitney U test. Size (mantle length), weight (total body mass) and sex were considered as factors of variation in the species with more than 8 individuals (*Loligo vulgaris*, *L. forbesi*, *Eledone cirrhosa* and *Sepia officinalis*) and their effects were checked using ANCOVA on log transformed values. The significance for statistical analyses was always set at $\alpha=0.05$.

3. Results

3.1. Levels of ²¹⁰Po in cephalopod digestive gland

The concentrations of ²¹⁰Po in the digestive gland of 11 species of cephalopods from the Bay of Biscay are given in [Table 1](#). Concentrations of ²¹⁰Po varied widely across the dataset, i.e.

from 188 Bq kg⁻¹ dw in the mesopelagic squid *Teutowenia megalops* to 11853 Bq kg⁻¹ dw in the neritic squid *Loligo vulgaris* (Table 1). On average, *L. vulgaris* contained the highest concentrations of ²¹⁰Po with 5720 ± 3606 Bq kg⁻¹ dw on average. More generally, the two species of loliginids (i.e. *L. forbesi* and *L. vulgaris*) showed the highest concentrations and the mesopelagic squids the lowest, whereas benthic and necto-benthic species had intermediate levels (Fig. 2). Considering the habitats (benthic vs pelagic and neritic vs oceanic), there was no significant difference (p>0.05).

3.2. Influence of size and sex on ²¹⁰Po concentrations

The variations of ²¹⁰Po concentrations in the digestive gland of cephalopods with size (mantle length) according to the sex were tested for the species with more than 8 individuals, i.e. *Loligo forbesi*, *L. vulgaris*, *Sepia officinalis* and *Eledone cirrhosa*. There was no difference due to sex in the four species. In these species, males and females when considered together to assess size effect. ²¹⁰Po concentrations significantly varied with size in only three species over four, i.e. *L. forbesi* (n=10), *L. vulgaris* (n=10) and *Eledone cirrhosa* (n=17), but not in *Sepia officinalis* (n=8; Fig. 3).

4. Discussion

The concentrations of ²¹⁰Po were considered in the digestive gland of several species of cephalopods from the Bay of Biscay as this organ plays a major role in the metabolism of trace elements in these molluscs, and of this natural radionuclide in particular (Heyraud and Cherry 1979; Smith et al. 1984; Finger and Smith 1987). As for some trace elements such as Ag, Cd, Cu and Zn (Miramand and Bentley 1992, Kojadinovic et al. 2011), the digestive gland of cephalopods contains over 90% of the whole-body burden of the ²¹⁰Po and the muscle tissues had only very low concentrations (Heyraud and Cherry 1979; Smith et al. 1984). But in contrast

to Ag, Cd, Cu and Zn that have high affinity to thiols groups of the metallothioneins in cephalopods (Bustamante et al. 2002b), ^{210}Po was rather associated with high molecular weight proteins ($> 70\text{kDa}$) in the digestive gland of the squid *Nototodarus gouldi* (Finger and Smith 1987). In this squid species, Smith et al. (1984) also pointed that there was no correlation between ^{210}Po and these metals, further suggesting a minor role of metallothioneins in its bioaccumulation. In contrast, ^{210}Po was shown correlate with Fe (Smith et al. 1984). Although there is only a limited number of studies that looked to molecular binding of ^{210}Po in marine organisms, it appears that ferritin in fish (i.e. the Atlantic mackerel *Scomber scombrus*), bivalves (i.e. the Pacific oyster *Crassostrea gigas*) and crustaceans (i.e., the lobster *Homarus gammarus*) plays an important role in the accumulation of ^{210}Po in the liver or digestive gland of these organisms (Durand et al. 1999, 2002), and the coaccumulation of both elements can support the binding of ^{210}Po to ferritin in the digestive gland of cephalopods too. However, these authors also showed that metallothioneins in fish and hemocyanin in lobster bind ^{210}Po to important extent. This highlights the need to further investigate specifically molecular binding of ^{210}Po in more details, considering that the radionuclide subcellular distribution may vary among cephalopods taxa (see Penicaud et al., 2017).

The efficient retention of ^{210}Po in this organ consistently lead to high concentrations in the different species investigated here and in those reported in the literature (Table 2). This review shows that all the species that have been analysed are species of commercial interest and consumed by humans, including octopus, cuttlefish and squids of the Ommastrephid and Loliginid families. To the best of our knowledge, our study is thus the first one to report values about the digestive gland ^{210}Po concentrations in a wide range of cephalopod species, considering benthic and pelagic species living in neritic and oceanic habitats, including mesopelagic species. The effect of the habitat on trace element concentrations in cephalopods

was significant for Cd and Hg in cephalopods from the same area, with benthic species having higher values than pelagic ones (Bustamante et al. 1998, 2006). In the present study, habitat did not clearly influence ^{210}Po concentrations, with pelagic species showing at the same time the highest (Loliginids) and the lowest (Ommastrephids and mesopelagic squids) values (Fig. 2). The present review also shows that ^{210}Po concentrations in the digestive gland of cephalopods varied over 4 order of magnitude, suggesting that the accumulation of ^{210}Po in this organ is likely influenced by biological factors, including the digestive physiology that differs between Ommastrephids and Loliginids (see Penicaud et al., 2017).

Among these biological factors, we have also considered the influence of sex and the size for the species with at least 8 individuals, i.e., the curled octopus *E. cirrhosa*, the common cuttlefish *S. officinalis*, the veined squid *L. forbesi* and the European squid *L. vulgaris*. We found no differences between ^{210}Po concentrations of males and females. This result is consistent with previous studies on the Japanese flying squid *Todarodes pacificus* and on the arrow squid *Nototodarus gouldi* (Smith et al. 1984; Waska et al. 2008). This suggests that the physiological mechanisms of ^{210}Po accumulation and elimination are not implied in the sexual maturation and gametogenesis.

In contrast to sex, size dramatically influenced ^{210}Po concentrations in the digestive gland of cephalopods (Fig. 3). Such a decrease with size has often been reported for a variety of marine organisms including crustaceans, molluscs and fish (Fowler 2011). As food is the dominant source of ^{210}Po in marine organisms (Heyraud and Cherry 1979; Cherry et al., 1989; Carvalho, 1990), this general decrease along size suggests a close relationship between ^{210}Po concentration in these marine organisms and their food ingestion rates (Fowler 2011). Also, it could result 1) from a dilution of ^{210}Po due to the fast growth of cephalopods and/or 2) from a switch in the diet along ontogeny as shown for some metals such as Cd and Hg (Chouvelon et al. 2011). To the best of our knowledge, there was a single study that considered the influence

of size on ^{210}Po concentrations in cephalopods (Heyraud et al. 1994). These authors reported an increase of ^{210}Po concentrations in whole squids from South Africa but they also considered unusual their own results as it was not following the decreasing paradigm. They suggested that a difference in diet between large and small squids could explain their unusual result. Variation of the diet with size in cephalopods is well documented (e.g., Nixon, 1987) and for instance, it results in higher Cd exposure of small sizes squids compared to adults, as the crustacean prey of the juveniles are Cd-enriched compared to Cd-poor fish (Penicaud et al. 2017). Thus, decreasing Cd concentrations with increasing body size was already reported for *L. forbesi* sampled in the Bay of Biscay and around the UK (Pierce et al. 2008; Chauvelon et al. 2011). In turn, ^{210}Po concentrations appeared to be higher in crustacean prey of squids than in fish (Heyraud et al. 1994, Carvalho 2011), which should lead to a decrease in ^{210}Po values as observed in the present study. However, in our sampling, the common cuttlefish showed no significant variation of ^{210}Po with size. Although this can be due to a limited number of individuals considered, a similar result has been reported for Cd in this species (Chauvelon et al. 2011), likely due to the opportunistic feeding behaviour of common cuttlefish. This plasticity is supported by investigations of their diet through stomach content analyses (Pinczon du Sel et al. 2000; Neves et al. 2009) as well as by the large variations in $\delta^{15}\text{N}$ values (a proxy of the trophic position) with size in this species, confirming that the common cuttlefish has a strong dietary plasticity at all stages of its life cycle (Chauvelon et al. 2011).

Cephalopods are preyed upon by many large predators such as tuna, swordfish, seabirds and marine mammals (Clark 1996, Croxall and Prince 1996, Smale 1996). The concentrations of Po in the muscle of the sperm whale (*Physeter macromphalus*) from the Azores, which feeds exclusively on mesopelagic cephalopods, were $5.0 \pm 0.2 \text{ Bq kg}^{-1} \text{ ww}$ (Carvalho 2011). This value is of the same order of magnitude as the one reported in the muscles of bigeye tuna ($3.0 \pm 0.1 \text{ Bq kg}^{-1}$) from Madeira Islands (Carvalho 2011), which feed on a variety of prey including

both epipelagic and mesopelagic organisms and includes fish and squids in its diet (Duffy et al. 2017). However, epipelagic marine mammals such as common and white sided dolphins *Delphinus delphis* and *Stenella coeruleoalba* have higher concentrations of ^{210}Po activity in their muscle tissue than sperm whales (Marta and Carvalho 2011), likely because they include small pelagic fish and Loliginids in their diet. To the best of our knowledge, there is no data on large predators from the Bay of Biscay while there is a large community of fish, seabirds and marine mammals foraging in this highly productive area. Future studies should investigate Po in the tissues of top predators, to determine if those feeding mainly on cephalopods, and particularly on the neritic ones such as delphinids (Lahaye et al. 2005) are at risk due to high exposure to this radioelement.

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References

- Belivermiş M, Kiliç Ö, Efe E, Sezer N, Gönülal O, Arslan Kaya TN (2019) Mercury and Po-210 in mollusc species in the island of Gökçeada in the north-eastern Aegean Sea: Bioaccumulation and risk assessment for human consumers. *Chemosphere*, 235: 876-884
- Boyle P, Rodhouse PG (2005) Cephalopods ecology and fisheries. Oxford: Blackwell Science.
- Bustamante P, Caurant F, Fowler SW, Miramand P (1998) Cephalopods as a vector for the transfer of cadmium to top marine predators in the North East Atlantic Ocean. *Science of the Total Environment*, 220: 71–80.

- Bustamante P, Teyssié JL, Fowler SW, Cotret O, Danis B, Miramand P, Warnau M (2002a) Biokinetics of zinc and cadmium accumulation and depuration at different stages in the life cycle of the cuttlefish *Sepia officinalis*. *Marine Ecology Progress Series*, 231: 167-177.
- Bustamante P, Cosson RP, Gallien, I, Caurant, F, Miramand P (2002b) Cadmium detoxification processes in the digestive gland of cephalopods in relation to accumulated cadmium concentrations. *Marine Environmental Research*, 53: 227-241.
- Bustamante P, Germain P, Leclerc G, Miramand P (2002c) Concentration and distribution of ^{210}Po in the tissues of the scallop *Chlamys varia* and the mussel *Mytilus edulis* from the coasts of Charente-Maritime (France). *Marine Pollution Bulletin*, 44(10): 997-1002.
- Bustamante P, Lahaye V, Durnez C, Churlaud C, Caurant F (2006) Total and organic Hg concentrations in cephalopods from the North East Atlantic waters: influence of geographical origin and feeding ecology. *Science of the Total Environment*, 368: 585-596.
- Carvalho FDP (1990) Contribution à l'étude du cycle du polonium-210 et du plomb-210 dans l'environnement (Doctoral dissertation, Nice).
- Carvalho FP (2011) Polonium (^{210}Po) and lead (^{210}Pb) in marine organisms and their transfer in marine food chains. *Journal of Environmental Radioactivity*, 102(5): 462-472.
- Carvalho FP, Fowler SW (1993) An experimental study on the bioaccumulation and turnover of polonium-210 and lead-210 in marine shrimp. *Marine Ecology Progress Series*, 102(1): 125-133
- Cherry RD, Heyraud M (1988) Lead-210 and polonium-210 in the world's oceans. *Inventories of Selected Radionuclides in the Oceans, IAEA-TECDOC-481 Vienna*, 139.
- Cherry RD, Heyraud M (1991) Polonium-210 and lead-210 in marine organisms: allometric relationships and their significance. In *Radionuclides in the study of marine processes* (pp. 309-318). Springer, Dordrecht.
- Cherry RD, Shannon LV (1974) The alpha radioactivity of marine organisms. *Atomic Energy Review*, 12(1): 3-45.
- Cherry RD, Heyraud M, Higgo JJW (1983) Polonium-210: its relative enrichment in the hepatopancreas of marine invertebrates. *Marine Ecology Progress Series*, 229-236.
- Chouvelon T, Spitz J, Cherel Y, Caurant F, Sirmel R, Mèndez-Fernandez P, Bustamante P (2011) Interspecific and ontogenic differences in $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values and Hg and Cd concentrations of cephalopods. *Marine Ecology Progress Series*, 433: 107-120.
- Clarke MR (1996) Cephalopods as prey. III. Cetaceans. *Philosophical Transactions of the Royal Society B*, 351: 1053-1065.
- Croxall JP, Prince PA (1996) Cephalopods as prey. I. Seabirds. *Philosophical Transactions of the Royal Society B* 351: 1023-1043.
- Danis B, Bustamante P, Cotret O, Teyssié J-L, Fowler SW, Warnau M (2005) Bioaccumulation of PCBs in the cuttlefish *Sepia officinalis* from sea water, sediment and food pathways. *Environmental Pollution*, 134: 113-122.
- Duffy LM, Kuhnert PM, Pethybridge HR, Young JW, Olson RJ, Logan JM, Goñi N, Romanov E, Allain V, Staudinger MD, Abecassis M, Choy CA, Hobday AJ, Simier M, Galván-Magaña F, Potier M, Ménard F (2017) Global trophic ecology of yellowfin, bigeye, and albacore tunas: Understanding predation on micronekton communities at ocean-basin scales. *Deep Sea Research Part II: Topical Studies in Oceanography, Future of oceanic animals in a changing ocean*, 140: 55-73.
- Durand JP, Carvalho FP, Goudard F, Pieri J, Fowler SW, Cotret O (1999) ^{210}Po binding to metallothioneins and ferritin in the liver of teleost marine fish. *Marine Ecology Progress Series*, 177: 189-196.

- Durand JP, Goudard F, Barbot C, Pieri J, Fowler SW, Cotret O (2002) Ferritin and hemocyanin: ^{210}Po molecular traps in marine fish, oyster and lobster. *Marine Ecology Progress Series*, 233: 199-205.
- Finger JM, Smith JD (1987) Molecular association of Cu, Zn, Cd and ^{210}Po in the digestive gland of the squid *Nototodarus gouldi*. *Marine Biology*, 95: 87-91.
- Fowler SW (2011) ^{210}Po in the marine environment with emphasis on its behaviour within the biosphere. *Journal of Environmental Radioactivity*, 102(5): 448-461.
- Guary J-C, Fowler SW (1982) Experimental studies on the biokinetics of plutonium and americium in the cephalopod *Octopus vulgaris*. *Marine Ecology Progress Series*, 7: 37-43.
- Heyraud M, Cherry RD (1979) Polonium-210 and lead-210 in marine food chains. *Marine Biology*, 52: 227-36.
- Heyraud M, Cherry RD, Oschadleus HD, Augustyn CJ, Cherry MI, Sealy JC (1994) Polonium-210 and Lead-210 in edible molluscs from near the Cape of Good Hope: Sources of variability in polonium-210 concentrations. *Journal of Environmental Radioactivity*, 24(3): 253-272.
- Khan MF, Wesley SG (2011a) Tissue distribution of ^{210}Po and ^{210}Pb in select marine species of the coast of Kudankulam, southern coast of Gulf of Mannar, India. *Environmental Monitoring and Assessment*, 175(1-4): 623-632.
- Khan MF, Godwin Wesley S (2011b) Bioaccumulation of ^{210}Po and ^{210}Pb in cephalopods collected from Kudankulam (Southeastern Coast of Gulf of Mannar, India) and assessment of dose in human beings. *Radiation Protection Dosimetry*, 147(3): 457-466.
- Kılıç Ö, Belivermiş M, Gönülal O, Sezer N, Carvalho FP (2018) ^{210}Po and ^{210}Pb in fish from northern Aegean Sea and radiation dose to fish consumers. *Journal of Radioanalytical and Nuclear Chemistry*, 318: 1189-1199.
- Klages NTW (1996) Cephalopods as prey. II. Seals. *Philosophical Transactions of the Royal Society B*, 351: 1045-1052.
- Kojadinovic J, Jackson CH, Chereil Y, Jackson GD, Bustamante P (2011) Multi-elemental concentrations in the tissues of the oceanic squid *Todarodes filippovae* from Tasmania and the Southern Indian Ocean. *Ecotoxicology and Environmental Safety*, 74(5): 1238-1249.
- Lahaye V, Bustamante P, Spitz J, Das K, Meynier L, Magnin V, Dabin W, Caurant F (2005) Long-term dietary segregation of common dolphins *Delphinus delphis* in the Bay of Biscay, determined using cadmium as an ecological tracer. *Marine Ecology Progress Series*, 305: 275-285.
- Lassalle G, Chouvelon T, Bustamante P, Niquil N (2014) An assessment of the trophic structure of the Bay of Biscay continental shelf food web: comparing estimates derived from an ecosystem model and isotopic data. *Progress in Oceanography*, 120: 205-215.
- Lorance P, Bertrand JA, Brind'Amour A, Rochet MJ, Trenkel VM (2009) Assessment of impacts from human activities on ecosystem components in the Bay of Biscay in the early 1990s. *Aquatic Living Resources*, 22: 409-431.
- Martin JH, Flegal AR (1975) High copper concentrations in squid livers in association with elevated levels of silver, cadmium, and zinc. *Marine Biology*, 30: 51-55.
- Malta M, Carvalho FP (2011) Radionuclides in marine mammals off the Portuguese coast. *Journal of Environmental Radioactivity*, 102: 473-478.
- Miramand P, Bentley D (1992) Concentration and distribution of heavy metals in tissues of two cephalopods, *Eledone cirrhosa* and *Sepia officinalis* from the French coast of the English Channel. *Marine Biology*, 114: 407-414.

- Neves A, Sequeira V, Vieira AR, Paiva R, Gordo LS (2009) Feeding habits of the cuttlefish *Sepia officinalis* during its life cycle in the Sado estuary (Portugal). *Hydrobiologia*, 636: 479–488.
- Nixon M (1987) Cephalopod diets. In: Boyle PR, editor. Cephalopod life cycles. Comparative Reviews. London: Academic Press; p. 201–19.
- OSPAR, 2010. Quality Status Report 2010. OSPAR (Oslo-Paris) Commission, London, UK.
- Penicaud V, Lacoue-Labarthe T, Bustamante P (2017) Metal bioaccumulation and detoxification processes in cephalopods: a review. *Environmental Research*, 155: 123–133.
- Pierce GJ, Stowasser G, Hastie LC, Bustamante P (2008) Geographic, seasonal and ontogenetic variation in cadmium and mercury concentrations in squid (Cephalopoda: Teuthoidea) from UK waters. *Ecotoxicology and Environmental Safety*, 70(3): 422–432.
- Pinczon du Sel G, Blanc A, Daguzan J (2000) The diet of the cuttlefish *Sepia officinalis* L. (Mollusca: Cephalopoda) during its life cycle in the Northern Bay of Biscay (France). *Aquatic Sciences*, 61: 167–178.
- Piñero-García F, Thomas R, Mantero J, Forssell-Aronsson E, Isaksson M (2022) Concentration of radionuclides in Swedish market basket and its radiological implications. *Food Control*, 133: 108658.
- Planque B, Lazure P, Jégou AM (2004) Detecting hydrological landscapes over the Bay of Biscay continental shelf in spring. *Climate Research*, 28: 41–52.
- Puillat I, Lazure P, Jégou AM, Lampert L, Miller PI (2004) Hydrographical variability on the French continental shelf in the Bay of Biscay, during the 1990s. *Continental Shelf Research*, 24: 1143–1163.
- Rodrigo AP, Costa PM (2017) The role of the cephalopod digestive gland in the storage and detoxification of marine pollutants. *Frontiers in Physiology*, 8: 232.
- Smale MJ (1996) Cephalopods as prey. IV. Fishes. *Philosophical Transactions of the Royal Society B*, 351: 1067–1081.
- Smith JD, Plues L, Heyraud M, Cherry RD (1984) Concentrations of the elements Ag, Al, Ca, Cd, Cu, Fe, Mg, Pb and Zn, and the radionuclides ^{210}Pb and ^{210}Po in the digestive gland of the squid *Nototodarus gouldi*. *Marine Environmental Research*, 13: 55–68.
- Strady E, Harmelin-Vivien M, Chiffoleau J-F, Veron A, Tronczynski J, Radakovitch O (2015) ^{210}Po and ^{210}Pb trophic transfer within the phytoplankton–zooplankton–anchovy/sardine food web: a case study from the Gulf of Lion (NW Mediterranean Sea). *Journal of Environmental Radioactivity*, 143: 141–151.
- Tanabe S, Tanaka H, Tatsukawa R (1984) Polychlorobiphenyls, DDT, and hexachlorocyclohexane isomers in the western North Pacific ecosystem. *Archives of Environmental Contamination and Toxicology*, 13: 731–738.
- Ueda T, Nakahara M, Ishii T, Suzuki Y, Suzuki H (1979) Amounts of trace elements in marine cephalopods. *Journal of Radioactivity Research*, 20: 338–342.
- Ueno D, Inoue S, Ikeda K, Tanaka H, Yamada H, Tanabe S (2003) Specific accumulation of polychlorinated biphenyls and organochlorine pesticides in Japanese common squid as a bioindicator. *Environmental Pollution*, 125: 227–235.
- Waska H, Kim S, Kim G, Kang MR, Kim GB (2008) Distribution patterns of chalcogens (S, Se, Te, and ^{210}Po) in various tissues of a squid, *Todarodes pacificus*. *Science of the Total Environment*, 392(2-3): 218–224.

Table 1. Characteristics of sampled cephalopods from the Bay of Biscay and the activity concentrations (Bq kg⁻¹ dw) of ²¹⁰Po in their digestive gland. Values are provided as mean ± SD and range (min-max). ND: Not determined.

Family Species	Sample Size	Mantle length (mm)		Fresh weight (g)		Sex	²¹⁰ Po	
		Mean ± SD	Range	Mean ± SD	Range		Mean ± SD	Range
Loliginidae								
<i>Loligo forbesi</i>	10	259 ± 73	160-440	499 ± 324	134-1287	3 ♂, 7 ♀	2551 ± 3144	342-10901
<i>L. vulgaris</i>	10	343 ± 105	190-470	920 ± 573	190-1790	6 ♂, 4 ♀	5720 ± 3606	1808-11853
Ommastrephidae								
<i>Todarodes sagittatus</i>	6	353 ± 67	225-420	1045 ± 462	220-1620	6 ♀	1735 ± 1786	532-5218
<i>Todaropsis eblanae</i>	4	151 ± 21	125-175	259 ± 134	147-450	2 ♂, 2 ♀	714 ± 258	332-880
Histioteuthidae								
<i>Histioteuthis reversa</i>	1		85		145	1 ♀		1206
Cranchidae								
<i>Teuthowenia megalops</i>	2		135-270		30-ND	ND		188-254
Sepiolidae								
<i>Rossia macrosoma</i>	1		56		52	1 ♀		3650
Sepiidae								
<i>S. officinalis</i>	8	183 ± 36	140-245	762 ± 406	300-1495	3 ♂, 5 ♀	1265 ± 783	366-2389
Octopodidae								
<i>Eledone cirrhosa</i>	17	90 ± 25	55-150	276 ± 248	60-1085	9 ♂, 8 ♀	1367 ± 384	823-2239
<i>Octopus vulgaris</i>	2		80-90		386-648	1 ♂, 1 ♀		1695-2561
<i>Octopus salutii</i>	1		60		92	ND		1585

Table 2. ^{210}Po concentrations in the digestive gland of cephalopods for selected families. All concentrations are presented in Bq kg^{-1} dw.

Family	Species	Location	Number	Mean \pm SD	Range	Reference
<i>Octopodidae</i>						
	<i>Eledone aldrovandii</i>	Mediterranean Sea	1	481		Heyraud and Cherry 1979
	<i>Eledone cirrhosa</i>	Bay of Biscay, Atlantic Ocean	17	1367 \pm 384	823-2239	Present study
	<i>Octopus vulgaris</i>	Mediterranean Sea	2	-	814-851	Heyraud and Cherry 1979
	<i>Octopus vulgaris</i>	Mediterranean Sea	4	1814 \pm 74		Heyraud and Cherry 1979
	<i>Octopus vulgaris</i>	Mediterranean Sea	1	2400*		Guarry et al. 1981
	<i>Octopus vulgaris</i>	Bay of Biscay, Atlantic Ocean	2	-	1695-2561	Present study
	<i>Octopus salutii</i>	Bay of Biscay, Atlantic Ocean	1	-	1585	Present study
<i>Sepiidae</i>						
	<i>Sepiella inermis</i>	South-eastern coast of India	32	745 \pm 52*	-	Khan & Wesley 2011a
	<i>Sepia officinalis</i>	Mediterranean Sea	3	1986 \pm 352	1628-2331	Heyraud and Cherry 1979
	<i>Sepia officinalis</i>	Bay of Biscay, Atlantic Ocean	8	1265 \pm 783	366-2389	Present study
	<i>Sepia pharaonis</i>	South-eastern coast of India	12	1232 \pm 48*	-	Khan & Wesley 2011b
<i>Loliginidae</i>						
	<i>Loligo forbesi</i>	Azores Islands, Atlantic Ocean	1	193*		Carvalho 2011
	<i>Loligo forbesi</i>	Bay of Biscay, Atlantic Ocean	10	2551 \pm 3144	342-10901	Present study
	<i>Loligo vulgaris</i>	Mediterranean Sea	5	3367 \pm 1702	1702-6068	Heyraud and Cherry 1979
	<i>Loligo vulgaris</i>	Atlantic Ocean	5	137 \pm 107	59-322	Heyraud and Cherry 1979
	<i>Loligo vulgaris</i>	Atlantic Ocean	4	2294 \pm 444	1813-2701	Heyraud and Cherry 1979
	<i>Loligo vulgaris</i>	Bay of Biscay, Atlantic Ocean	10	5720 \pm 3606	1808-11853	Present study
	<i>Uroteuthis duvauceli</i>	South-eastern coast of India	22	1799 \pm 69*	-	Khan & Wesley 2011
	<i>Uroteuthis duvauceli</i>	South-eastern coast of India	25	825 \pm 69*	-	Khan & Wesley 2011
<i>Ommastrephidae</i>						
	<i>Nototodarus gouldi</i>	Bass Strait, Australia	26	9600 \pm 4600	4800-24200	Smith et al. 1984
	<i>Nototodarus gouldi</i>	Port Philip Bay, Australia	6	2900 \pm 1600	1700-5600	Finger & Smith 1987
	<i>Todarodes pacificus</i>	Korean East coast, Japan Sea	10	1208 \pm 269	695-1819	Waska et al. 2008
	<i>Todarodes sagittatus</i>	Bay of Biscay, Atlantic Ocean	6	1735 \pm 1786	532-5218	Present study
	<i>Todaropsis eblanae</i>	Bay of Biscay, Atlantic Ocean	4	714 \pm 258	332-880	Present study

* converted from wet weight using a factor 4.1 according to Heyraud and Cherry 1979

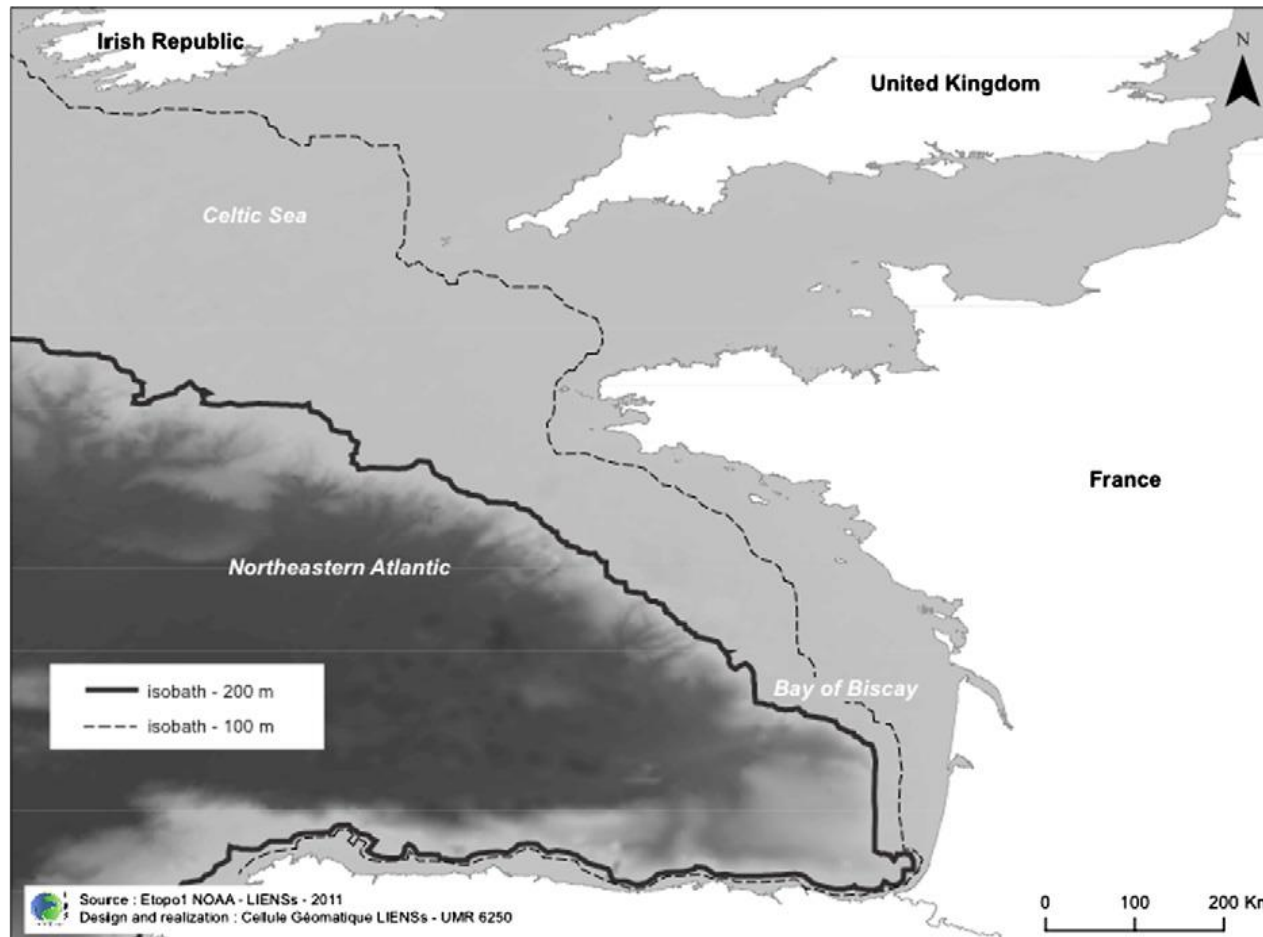


Figure 1. Map of the study area.

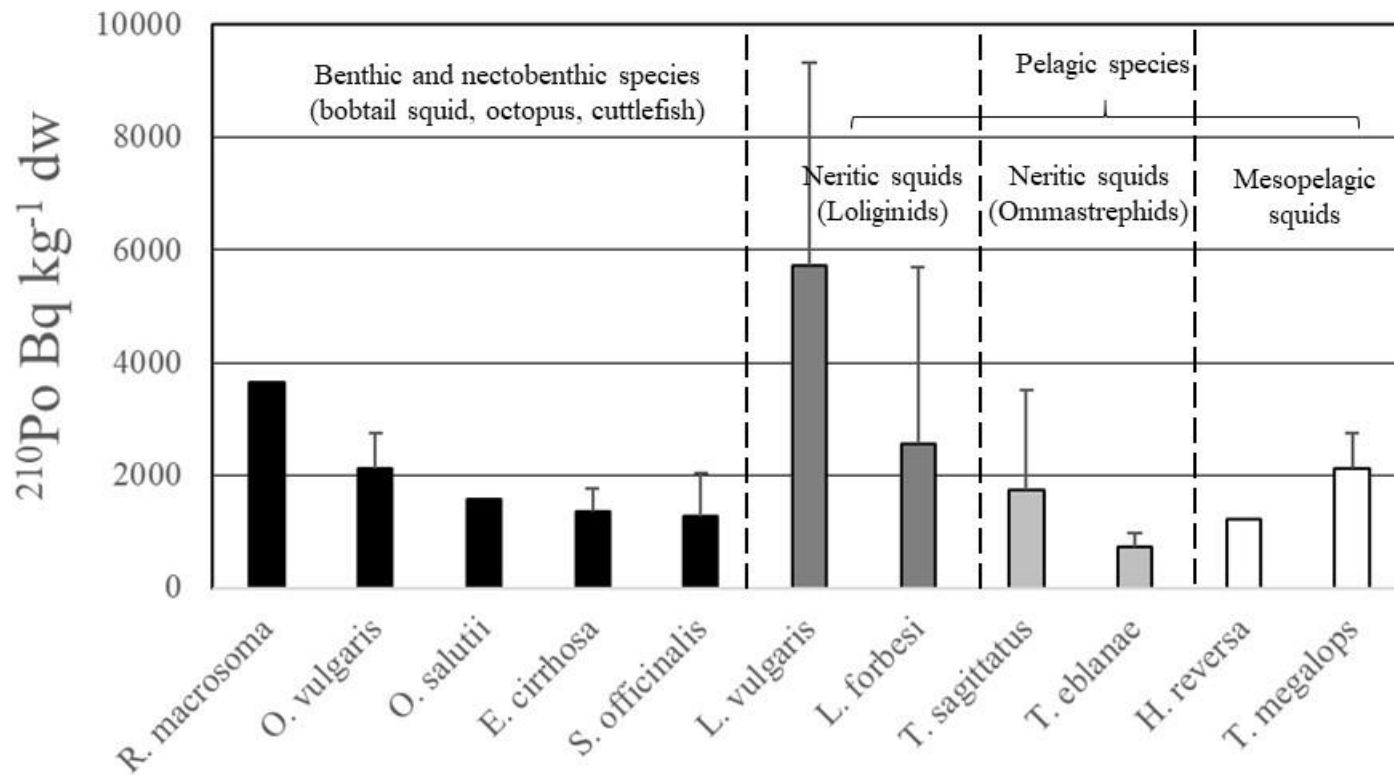


Figure 2. Concentrations of ^{210}Po (Bq kg⁻¹ dw) in the digestive gland of 11 species of cephalopods from the Bay of Biscay, North Atlantic Ocean. Species are classified by habitats (benthic and nectobenthic, neritic, pelagic and mesopelagic species) and by family for neritic squids (Loliginids and Ommastrephids).

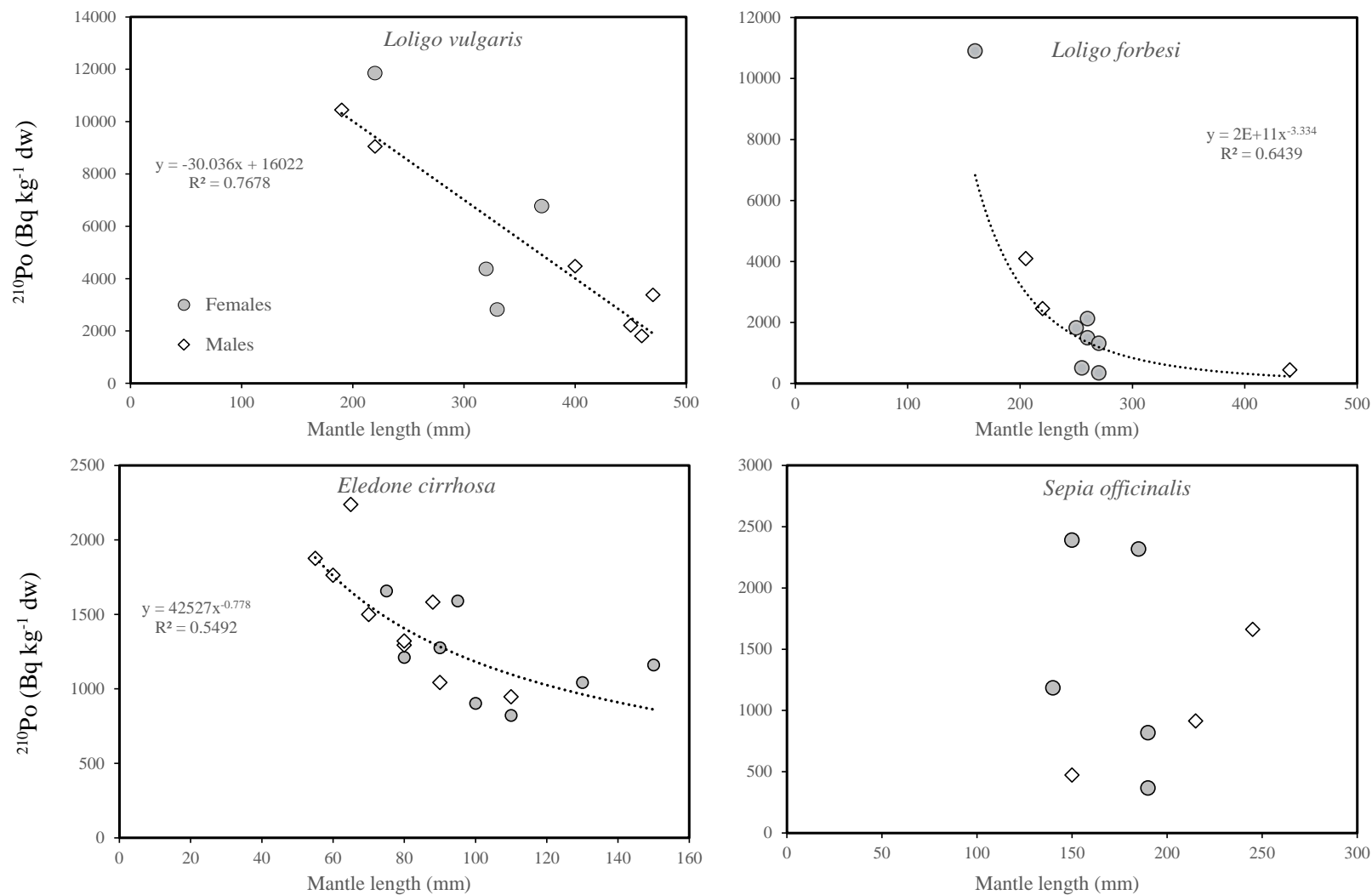


Figure 3. Relationship between ^{210}Po concentrations (Bq kg⁻¹ dw) in the digestive gland and body size (mantle length in mm) in 4 species of cephalopods from the Bay of Biscay, North Atlantic Ocean. Males and females are represented in white diamonds and grey circles, respectively.