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Biodistribution of ^{210}Po in seafood and risk assessment for consumers in Sweden

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ABSTRACT

Seafood consumption per capita, in Sweden, is larger than World and European average. Although, 36% of Swedes consume seafood meals at least two times per week, the Swedish National Food Agency advises the necessity to increase this ratio. Seafood is one of the main entrances of ^{210}Po in the human food chain. Due to the high radiotoxicity, the intake of ^{210}Po plays an important role in the human health, even in extremely small quantities. In this study, 114 seafood samples representing 52 different marine species were analyzed. The biodistribution of ^{210}Po in seafood species were not uniformly distributed being higher in digestive system and gonads, and lower in seafood muscle. The activity concentration of ^{210}Po in fish ranged between 0.01 and 26 Bq/kg with an average value of 4 Bq/kg, whereas in shellfish fluctuated between 0.1 and 239 Bq/kg, with a mean concentration of 18 Bq/kg. In general, the activity concentration of ^{210}Po in processed products were lower than fresh samples due to the decay of ^{210}Po from seafood capture to purchase. However, in boiled seafood such as Norway Lobster, with short elapsed time from collection to purchase, the boil samples presented higher activity concentration of ^{210}Po than fresh products. The results of the study showed that the annual intake of ^{210}Po via seafood consumption in Sweden exponentially increased by age and it was slightly higher in males than females. As a result, the annual committed dose ranged from 60 to 154 μSv, with an average value of 103 ± 31 μSv, being controlled by fish consumption below 14 years old and by seafood consumption above 14 years old. Finally, the committed effective dose could increase up to 479 μSv/y for population group with higher seafood consumption.

1. Introduction

The entrance of radionuclides into the food chain is a problem for both humans and animals which can lead to adverse health effects. For humans, the ingestion of radionuclides contributes to approximately 12% of the annual effective dose received by population, 2.4 mSv (UNSCEAR, 2000). The effective dose largely depends on the food habits as well as the origin of the food. Most of the ingestion dose received by humans is due to the uptake of naturally occurring radionuclides from the progeny of uranium and thorium series (UNSCEAR, 2000). The radiotoxicity of the radionuclides with the major impact in food consumption decreases in the order ^{210}Po > ^{228}Ra > ^{210}Pb > ^{226}Ra > ^{234}U > ^{238}U > ^{224}Ra > ^{235}U (UNSCEAR, 2000).

Dietary habits change as a consequence of population mixing and due to varying availability of food products will also affect the ingested radionuclides. In Sweden, the consumption of seafood has increased during the last decades (Martin et al., 2016; SCB, 2015). In 2019, approximately 123 777 tonnes of edible seafood were available in Sweden representing a per capita consumption of 27 kg (RISE, 2021). The worldwide seafood consumption per capita in 2019 was 20.5 kg. Swedish National Food Agency advices eating fish and shellfish at least two or three times per week due to the health benefits; however, RISE reports concluded that "*Swedish consumption of seafood still does not reach the dietary advice of the Swedish National Food Agency*" (RISE, 2021).

Seafood plays an important role in the entrance of ^{210}Po in the human food chain since it is strongly accumulated in the marine biota (IAEA, 2017; Struminska-Parulska et al., 2013). For that reason, in countries with high consumption of seafood, ^{210}Po is the main radionuclide responsible for the committed effective dose from food, representing approximately 80% of the total (Diaz-Frances et al., 2016; Komperød et al., 2019; Piñero-García et al., 2022). ^{210}Po is an alpha emitter (5.3 MeV) and one of the most radiotoxic radionuclides,

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Table 1
Committed effective dose coefficient of ^{210}Po (μSv/Bq) for internal exposure via ingestion (food or water). Data is given as mean value and P95 of the annual consumption (kg/y) of fish and shellfish for different age groups, and also separated by gender.

Age	$e_{^{210}Po}$ (μSv/Bq)	Woman				Man				All			
		Fish		Shellfish		Fish		Shellfish		Fish		Shellfish	
		mean	P95	mean	P95	mean	P95	mean	P95	mean	P95	mean	P95
4 y	4.4	14	46	2	5	16	55	2	6	15	52	2	6
8–11 y	2.6	20	90	2	10	18	99	1	7	19	95	2	9
14 y	1.6	20	97	2	15	30	130	2	10	25	114	2	13
17 y	1.2	12	65	3	17	22	108	5	25	17	87	4	21
18–30 y		19	55	5	14	25	81	6	20	22	69	5	17
31–44 y		26	82	7	20	27	99	7	25	26	86	7	22
45–64 y		32	91	8	23	35	115	9	29	34	103	8	26
65–80 y		38	98	10	25	47	110	12	27	42	102	11	26
> 18 y		30	86	7	21	34	107	9	27	31	95	8	24

moreover, it can pass through biological membranes favouring its bioaccumulation due to its affinity to proteins (IAEA, 2017). Consequently, ^{210}Po plays an important role in the human health, even in extremely small quantities (trace levels) (Diaz-Frances et al., 2016).

Hence, this study aims to analyse ^{210}Po in fresh seafood as well as various processed seafood (dried, brined, canned and frozen), and also to analyse the biokinetics of ^{210}Po by studying different organs and tissues. The obtained results will be valuable for conducting a more realistic dose assessment, since not all parts of seafood are eaten. Furthermore, the processing of food could further dilute or concentrate the activity concentration of ^{210}Po, thus, this will also be studied.

2. Materials and methods

Seafood samples representing 52 different marine species were purchased from various vendors available for the Swedish population; a total of 114 samples were analyzed. Whenever possible fish and shellfish were acquired in their fresh, frozen, canned, brined and/or dried forms, considering also different origins and brands. For each sample, approximately 0.5–4 kg were collected, sampling at least ten individuals per marine species and three cans of the same brand per canned sample. After the collection, the individuals of each marine species as well as the cans or processed products of the same brand were combined as individual samples. In general, the determination of ^{210}Po was carried out in meat samples, therefore, non-consumed parts, such as shells from shrimps or bones and skins from fishes, were removed. However, in fresh samples of Atlantic mackerel (*Scomber scombrus*, fish), Norway lobster (*Nephrops norvegicus*, crustacean) and Great Atlantic scallop (*Pecten maximus* mollusk), organs and tissues were dissected to study the biodistribution of ^{210}Po in fish and shellfish. All the samples were weighed, and oven dried at 80 °C to minimize the loss of polonium during the drying (IAEA, 2017). Moreover, the moisture content was determined.

The determination of ^{210}Po was done 7 days after the sampling date at the latest to reflect the real intake of ^{210}Po by seafood consumption and to minimize the ^{210}Po decay and in-growth of ^{210}Po from ^{210}Pb. ^{210}Po determination by alpha spectrometry was performed in an aliquot of 0.25–4 g of dried sample. To assess the chemical yield and calculate the activity concentration of ^{210}Po, the samples were spiked with ^{209}Po (ca. 50 mBq). The internal standard was prepared with a certified standard solution of ^{209}Po (Eckert & Ziegler® Isotope Products, USA, reference number 1895–42). The samples were digested using an Ethos Easy microwave digestion system (Milestone S.R.L., Italy). For that, 5 mL of nitric acid (HNO_3, 65%, Fisher Scientific™; UK), 5 mL of distilled water and 2 mL of hydrogen peroxide (H_2O_2, 35%, Chem Lab nv, Belgium) were used. The microwave acid digestion procedure was 15 min of ramp time, 15 min of step time and 1800 W. After the acid digestion, the samples were diluted in 1 L with distilled water. Then, polonium was concentrated by co-precipitation of iron (III) using 30 mg of Fe^{3+} carrier and adjusting the pH between 8 and 9 with ammonia solution (25%, Fisher Scientific™; UK) to get the hydroxide precipitation ($Fe(OH)_3$). The iron carrier solution was prepared by iron (III) chloride hexahydrate (Fisher Scientific™; UK). The precipitate was collected, dried, and later dissolved in 30 mL of HCl (1.5M, prepared by HCl, 37% Fisher Scientific™; UK) together with 150 mg of L-ascorbic acid (Sigma-Aldrich, USA) to carry out the self-deposition of Po on copper discs at 80 °C during 4h. The activity concentration of ^{210}Po on the copper discs were measured by alpha-particle spectrometry. Further details regarding the radiometric characterization as well as the validation and the analytical quality control of the radiometric determination are found in previous publications (Piñero-García et al., 2021, 2022).

Finally, assessment of the committed effective dose from seafood ingestion during one year for different gender and age groups in the Swedish population was calculated using the effective dose coefficient for internal exposure via ingestion from International Commission on Radiological Protection publication 119 (ICRP, 2012), and the following equation:

$$E\ (\mu Sv / y) = \sum A_{^{210}Po} \bullet M_i \bullet e_{^{210}Po}$$

$$\text{where} \begin{cases} A_{^{210}Po}\ is\ \text{the activity concentration of } ^{210}Po \text{ refers to fresh weigth } \left[\mathrm{Bq/kg}_{f.w.}\right] \\ M_i\ is\ the\ annual\ consumption\ of\ seafood\ for\ Swedish\ population\ group\ (kg/y) \\ e_i\ is\ \text{the effective dose coefficient of } ^{210}Po\ [\mu\mathrm{Sv/Bq}] \end{cases}$$

Table 1 shows the effective dose coefficient of ^{210}Po (ICRP, 2012) together with the average and the 95th percentile (P95) of the annual consumption of fish and shellfish for the different age groups and divided for women, men and the entire population (Livsmedelsverket, 2006, 2012, 2018).

Table 2
Activity concentration of ^{210}Po in seafood (mBq/kg$_{f.w.}$). All activity concentrations refer to fresh weight and they are shown with an uncertainty of one standard deviation (coverage factor k = 1). Additionally, Latin name and information about the type of samples are provided. Processed samples refer to samples in canned or brined.

Sample	Latin name	Product type	^{210}Po (mBq/kg$_{f.w.}$)
Algae: Kelp	*Laminaria angustata*	Dried	1980 ± 191
Algae: Kombu	*Saccharina japonica*	Dried	787 ± 100
Algae: Seasoned laver	*Porphyra*	Dried	903 ± 109
Atlantic cod	*Gadus morhua*	Fresh	169 ± 9
Atlantic cod	*Gadus morhua*	Fresh	1090 ± 16
Atlantic cod	*Gadus morhua*	Fresh	122 ± 4
Atlantic cod	*Gadus morhua*	Fresh	1370 ± 45
Atlantic herring	*Clupea harengus*	Fresh	1050 ± 8
Atlantic herring	*Clupea harengus*	Fresh	961 ± 12
Atlantic herring	*Clupea harengus*	Fresh	923 ± 27
Atlantic herring	*Clupea harengus*	Processed	<100
Atlantic herring	*Clupea harengus*	Processed	116 ± 23
Atlantic herring	*Clupea harengus*	Processed	167 ± 26
Baltic herring	*Clupea harengus membras*	Fresh	2350 ± 105
Atlantic mackerel	*Scomber scombrus*	Frozen	643 ± 91
Atlantic mackerel	*Scomber scombrus*	Processed	<100
Atlantic mackerel	*Scomber scombrus*	Processed	5340 ± 242
Atlantic mackerel	*Scomberscombrus*	Processed	246 ± 28
Atlantic mackerel	*Scomber scombrus*	Fresh	1930 ± 83
Atlantic salmon (farm)[a]	*Salmo salar*	Fresh	13 ± 1
Atlantic salmon (wild)[b]	*Salmo salar*	Fresh	170 ± 7
Caviar: Atlantic cod	*Gadus morhua*	Processed	203 ± 48
Caviar: European anchovy	*Engraulis encrasicolus*	Processed	2060 ± 248
Caviar: Mackerel Atlantic	*Scomber scombrus*	Processed	221 ± 39
Caviar: Tuna Skipjack	*Katsuwonus pelamis*	Processed	2150 ± 206
Char	*Salvelinus* spp	Fresh	<100
Clam: Common edible cockle	*Cerastoderma edule*	Fresh	10700 ± 467
Clam: Japanese carpet shell	*Ruditapes philippinarum*	Fresh	239000 ± 6910
Clam: Japanese carpet shell	*Ruditapes philippinarum*	Fresh	124000 ± 6370
Clam: White clam	*Meretrix lyrata*	Frozen	2070 ± 251
Crab: Brown crab	*Cancer pagurus*	Boiled	19400 ± 1130
Crab: Brown crab	*Cancer pagurus*	Boiled	5270 ± 395
Crab: Red King crab (stick)	*Paralithodes camtschaticus*	Boiled	735 ± 97
Crab: Red swimming crab	*Portunus haanii*	Processed	638 ± 48
European anchovy	*Engraulis encrasicolus*	Processed	26000 ± 1010
European anchovy	*Engraulis encrasicolus*	Processed	10900 ± 784
European hake	*Merluccius merluccius*	Fresh	1150 ± 46
European pilchard (sardine)	*Sardina pilchardus*	Processed	794 ± 63
European pilchard (sardine)	*Sardina pilchardus*	Processed	17300 ± 713
European pilchard (sardine)	*Sardina pilchardus*	Processed	851 ± 68
European plaice	*Pleuronectes platessa*	Fresh	1300 ± 20
European plaice	*Pleuronectes platessa*	Fresh	246 ± 5
European plaice	*Pleuronectes platessa*	Fresh	1370 ± 16
European plaice	*Pleuronectes platessa*	Fresh	1230 ± 24
European plaice	*Pleuronectes platessa*	Fresh	1300 ± 33
European sprat	*Sprattus sprattus*	Processed	968 ± 154
European sprat	*Sprattus sprattus*	Processed	856 ± 141
European sprat	*Sprattus sprattus*	Processed	1440 ± 228
European sprat	*Sprattus sprattus*	Processed	1230 ± 84
European sprat	*Sprattus sprattus*	Processed	328 ± 28
Greater weever	*Trachinidae draco*	Fresh	<100
Lake herring	*Coregonus artedi*	Frozen	<100
Lake whitefish	*Coregonus clupeaformis*	Frozen	871 ± 56
Mussel: Blue mussel	*Mytilus edulis*	Frozen	36300 ± 1610
Mussel: Blue mussel	*Mytilus edulis*	Frozen	35800 ± 1800
Mussel: Blue mussel	*Mytilus edulis*	Processed	52900 ± 3940

Table 2 (*continued*)

Sample	Latin name	Product type	^{210}Po (mBq/kg$_{f.w.}$)
Mussel: Blue mussel	*Mytilus edulis*	Processed	15000 ± 871
Mussel: Blue mussel	*Mytilus edulis*	Fresh	73000 ± 5480
Mussel: Blue mussel	*Mytilus edulis*	Fresh	36500 ± 1220
Mussel: Blue mussel	*Mytilus edulis*	Fresh	10000 ± 975
Mussel: Chilean mussel	*Mytilus chilensis*	Frozen	11700 ± 577
Mussel: Chilean mussel	*Mytilus chilensis*	Frozen	4670 ± 389
Mussel: Chilean mussel	*Mytilus chilensis*	Processed	11400 ± 790
Mussel: Chilean mussel	*Mytilus chilensis*	Processed	3630 ± 353
Mussel: New Zealand mussel	*Perna canaliculus*	Frozen	6080 ± 434
Norway lobster	*Nephrops norvegicus*	Fresh	1950 ± 409
Norway lobster	*Nephrops norvegicus*	Boiled	5900 ± 436
Norway lobster	*Nephrops norvegicus*	Boiled	4370 ± 314
Octopus	*Octopus vulgaris*	Processed	1120 ± 170
Oyster: Pacific cupped oyster	*Crassostrea gigas*	Fresh	44300 ± 1770
Oyster: Pacific cupped oyster	*Crassostrea gigas*	Fresh	27600 ± 1300
Oyster: European flat oyster	*Ostrea edulis*	Fresh	8360 ± 760
Pike-perch	*Sander lucioperca*	Fresh	496 ± 25
Pike-perch	*Sander lucioperca*	Fresh	236 ± 9
Razor shells	*Solenidae*	Processed	2460 ± 221
Red swamp crawfish	*Procambarus clarkii*	Processed	602 ± 94
Red swamp crawfish	*Procambarus clarkii*	Processed	367 ± 40
Red swamp crawfish	*Procambarus clarkii*	Processed	100 ± 17
Red swamp crawfish	*Procambarus clarkii*	Processed	576 ± 54
Roe: Atlantic cod	*Gadus morhua*	Fresh	676 ± 124
Roe: Atlantic herring	*Clupea harengus*	Processed	259 ± 27
Roe: Lumpfish	*Cyclopterus lumpus*	Processed	<100
Saithe	*Pollachius virens*	Fresh	390 ± 29
Saithe	*Pollachius virens*	Fresh	562 ± 18
Scallop: American sea scallop	*Placopecten magellanicus*	Frozen	743 ± 51
Scallop: American sea scallop	*Placopecten magellanicus*	Frozen	558 ± 50
Scallop: Great Atlantic scallop	*Pecten maximus*	Fresh	5970 ± 428
Scallop: Yesso scallop	*Patinopecten yessoensis*	Fresh	799 ± 62
Shrimp: Giant tiger prawn	*Penaeus monodon*	Frozen	<100
Shrimp: Northern prawn	*Padalus borealis*	Frozen	<100
Shrimp: Northern prawn	*Padalus borealis*	Frozen	6600 ± 257
Shrimp: Northern prawn	*Padalus borealis*	Frozen	200 ± 26
Shrimp: Northern prawn	*Padalus borealis*	Frozen	2150 ± 115
Shrimp: Northern prawn	*Padalus borealis*	Frozen	<100
Shrimp: Northern prawn	*Padalus borealis*	Frozen	<100
Shrimp: Northern prawn	*Padalus borealis*	Frozen	443 ± 38
Shrimp: Northern prawn	*Padalus borealis*	Processed	604 ± 146
Shrimp: Northern prawn	*Padalus borealis*	Processed	2570 ± 175
Shrimp: Northern prawn	*Pandalus borealis*	Boiled	29800 ± 951
Shrimp: Whiteleg shrimp	*Penaeus vannamei*	Frozen	<100
Shrimp: Whiteleg shrimp	*Penaeus vannamei*	Frozen	<100
Squid: European flying squid	*Todarodes sagittatus*	Fresh	592 ± 53
Squid: Longfin squid	*Loligo* spp	Processed	929 ± 158
Squid: Northern shortfin squid	*Illex illecebrosus*	Frozen	244 ± 17
Surimi	*Surimi*	Processed	<100
Tuna	*Thunnus obesus*	Fresh	6210 ± 482
Tuna Skipjack	*Katsuwonus pelamis*	Processed	9330 ± 439
Tuna Skipjack	*Katsuwonus pelamis*	Processed	14200 ± 623
Tuna Skipjack	*Katsuwonus pelamis*	Processed	16600 ± 685

(*continued on next page*)

Table 2 (*continued*)

Sample	Latin name	Product type	^{210}Po (mBq/kg$_{f.w.}$)
Tuna Skipjack	*Katsuwonus pelamis*	Processed	5650 ± 255
Tuna Skipjack	*Katsuwonus pelamis*	Processed	22100 ± 1190
Tuna Skipjack	*Katsuwonus pelamis*	Processed	5310 ± 336
Tuna (longfin)/ albacore	*Thunnus alalunga*	Frozen	390 ± 61
Tuna (longfin)/ albacore	*Thunnus alalunga*	Processed	426 ± 160
Tuna (Yellowfin)	*Thunnus albacares*	Fresh	<100
Wakame	*Undaria pinnatifida*	Processed	778 ± 120

[a] (Heldal et al., 2019).
[b] (Komperød et al., 2019).

3. Results and discussions

3.1. Activity concentration of ^{210}Po in seafood

Table 2 shows the activity concentration of ^{210}Po in seafood samples, whereas Fig. 1 summarizes the results of the activity concentration of ^{210}Po (mean ± SD) clustering fish and shellfish samples by group of species (clam, oyster, lake fish, etc.) or product (caviar). For that, the average values presented in Fig. 1 were calculated considering the samples of each cluster with activity concentration higher than MDA and presented together with the standard deviation of each cluster. The activity concentration of ^{210}Po was higher than the minimum detectable activity (MDA, 100 mBq/kg$_{f.w.}$) in 88% of the seafood samples analyzed. The lowest activity concentration of ^{210}Po was found in farm Atlantic Salmon from Norway (13 ± 1 mBq/kg$_{f.w.}$) (Heldal et al., 2019), whereas the highest activity concentration of ^{210}Po was measured in a clam sample of Japanese carpet shell (239 ± 14 Bq/kg$_{f.w.}$) dredged by hand in the North zone of the Bay of Biscay (FAO27.VIII sub-A). On one hand, the activity concentration of ^{210}Po in shellfish ranged between 0.1 Bq/kg and 239 Bq/kg$_{f.w.}$, with an average value of 18 Bq/kg$_{f.w.}$. On the other hand, the activity concentration of ^{210}Po in fish was between 0.01 and 26 Bq/kg$_{f.w.}$ with a mean value of 4 Bq/kg$_{f.w.}$.

High activity concentration of ^{210}Po was detected in suspension feeders such as Japanese carpet shell (*Ruditapes philippinarum*), blue mussel (*Mytilus edulis*) and oyster (*Crassostrea gigas*), and zooplankton feeders such as European anchovy (*Engraulis encrasicolus*), sardine (*Sardina pilchardus*), Baltic herring (*Clupea harengus membras*) and Atlantic mackerel (*Scomber scombrus*). Those results demonstrate that ^{210}Po bioaccumulation in marine environment is higher in primary producers and primary consumers than top predators (Bustamante et al., 2002; Carvalho, 2011; Hansen et al., 2022). However, one exception, is the high ^{210}Po concentrations found in different samples of tuna, both fresh bigeye tuna (*Thunnus obesus*) and canned skipjack tuna (*Katsuwonus pelamis*). Tuna is a top predator with a diet rich in ^{210}Po, including small fish such as mackerel and herrings, as well as zooplankton. Therefore, their diet is rich in ^{210}Po and their metabolism favours the bioaccumulation of ^{210}Po in tuna organs and tissues (Carvalho, 2011; IAEA, 2017).

In general, the activity concentration of ^{210}Po in processed products such as frozen seafood was lower than ^{210}Po concentration in fresh products of the same species, probably due to ^{210}Po decay as a result of the elapsed time from capture to purchase of the processed products. However, boiled samples, with low elapsed time from capture to purchase, such as Norway lobster presented with higher ^{210}Po activity concentration than fresh samples, highlighting that even if polonium is volatile, cooking process could also increase the concentration of ^{210}Po due to polonium remains bound to the tissues or organs and the reduction of water content. In addition, the results of boiled Northern prawn also pointed out this trend, having the highest activity concentration of ^{210}Po detected in processed shrimps.

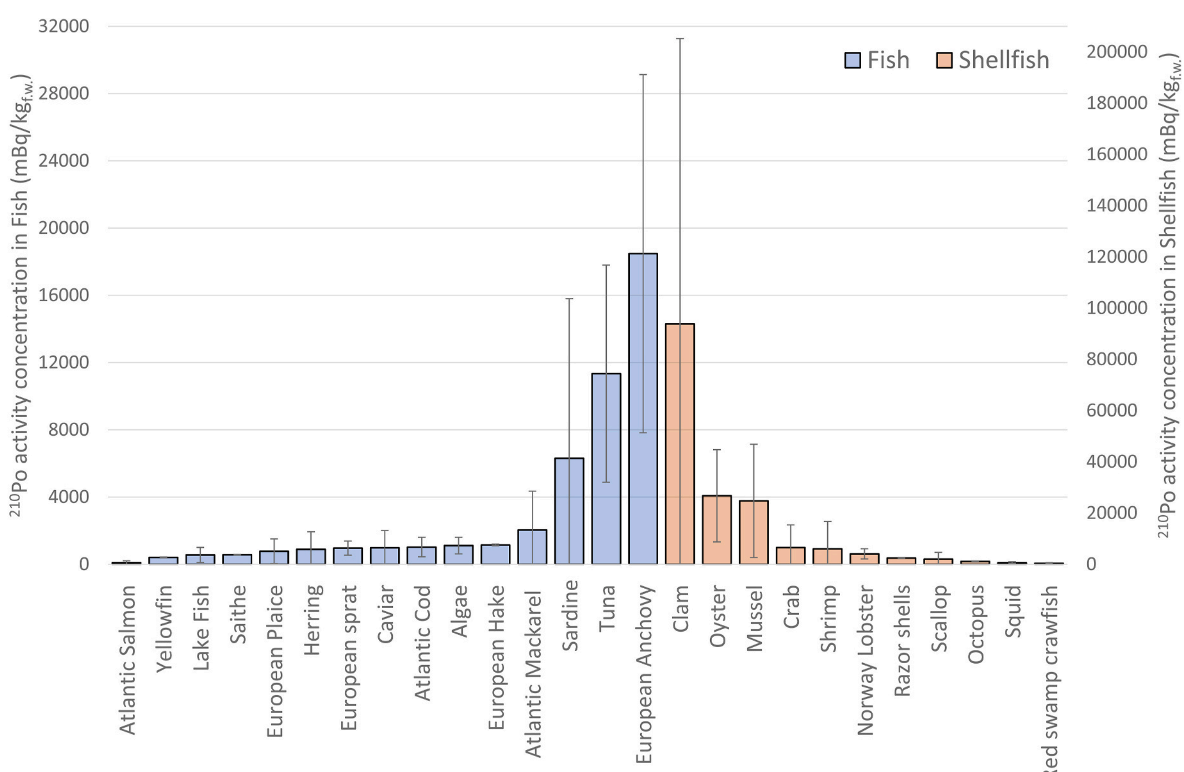

Fig. 1. Average activity concentration of ^{210}Po in fish and seafood species. Fish and shellfish samples have been clustered by group of species or products, considering those samples with activity concentration of ^{210}Po above MDA. Data is given by mean value together with the SD of each cluster. For those clusters represented by just one sample (Saithe, European hake, Razor shells and Octopus), Figure shows the activity concentration of ^{210}Po in the sample with an uncertainty of one standard deviation (coverage factor k = 1).

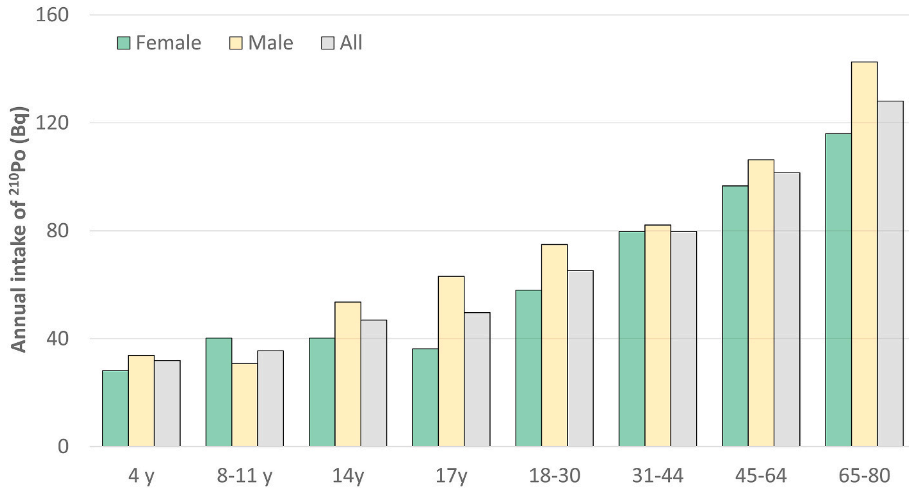

Fig. 2. Annual intake of ^{210}Po via seafood consumption in Sweden, given by age and gender.

Table 3
Activity concentration of ^{210}Po detected in different dissected organs and tissues from Atlantic mackerel, Great Atlantic scallop and Norway lobster (mBq/kg$_{f.w.}$). All activity concentrations refer to fresh weight and they are shown with an uncertainty of one standard deviation (coverage factor k = 1).

	^{210}Po (mBq/kg$_{f.w.}$)
Atlantic mackerel (Scomber scombrus)	
Bones	4290 ± 257
Digestive system	76900 ± 7220
Gills	2930 ± 110
Gonads	110000 ± 11100
Heart	22100 ± 2150
Liver	79500 ± 4620
Muscle	1930 ± 83
Kidney	29500 ± 2730
Skin	6550 ± 565
Norway lobster (Nephrops norvegicus)	
Gills	9170 ± 951
Gonads	450000 ± 39700
Heart	4270 ± 2620
Hepatopancreas	6340 ± 2770
Intestines	5100 ± 2260
Muscle	2110 ± 410
Stomach	26000 ± 2980
Great Atlantic scallop (Pecten maximus)	
Adductor muscle	5970 ± 855
Coral	18500 ± 2950
Gills	36300 ± 4280
Mantle skirts	6570 ± 978
Visceral mass	160000 ± 18200

Fig. 2 displays the average values of the annual intake of ^{210}Po in Sweden by age and gender. ^{210}Po intake was calculated based on the mean values of seafood consumption in Sweden (Table 1) and the average value of the activity concentration of ^{210}Po in fish and shellfish. The annual intake of ^{210}Po from seafood ingestion varied from 28 Bq to 348 Bq with a mean value of 156 ± 102 Bq. The annual intake of ^{210}Po from seafood in Sweden exponentially increased by age and it was slightly higher in males than females (Fig. 2). In Europe, the estimated annual intake of ^{210}Po from ingestion of water and food is around 14–135 Bq (IAEA, 2017). Furthermore, UNSCEAR reported a worldwide annual intake of ^{210}Po from ingestion (both water and food) in infants (1 year old), children (10 year old) and adults (>18 year old) of 21, 39 and 58 Bq, respectively (IAEA, 2017; UNSCEAR, 2000). The results presented in the current investigation showed that children in Sweden reached the worldwide annual intake of ^{210}Po only considering seafood ingestion. In addition, the same tendency occurred in adult population, however, with higher yearly intake of about 88 ± 26 Bq. Really, the annual intake of ^{210}Po is higher in countries or regions with high consumption of seafood such as Japan (225 Bq), Seville (Spain, 375 Bq) or Portugal (400 Bq) (Diaz-Frances et al., 2016). Though, in some regions with low consumption of seafood, high intake of ^{210}Po had been reported such as Northern Sweden (Lapland, 900 Bq) or Northwest Canada (1200 Bq) as a result of the high bioaccumulation of ^{210}Po in the typical food chain of these territories: Non-vascular plants (Lichen, mosses): Reindeer: Human (Diaz-Frances et al., 2016).

3.2. *Biokinetic distribution of ^{210}Po in seafood*

Table 3 shows the results of the activity concentration of ^{210}Po in tissues and organs of seafood species analyzed: Atlantic mackerel (*Scomber scombrus*, fish), Norway lobster (*Nephrops norvegicus*,

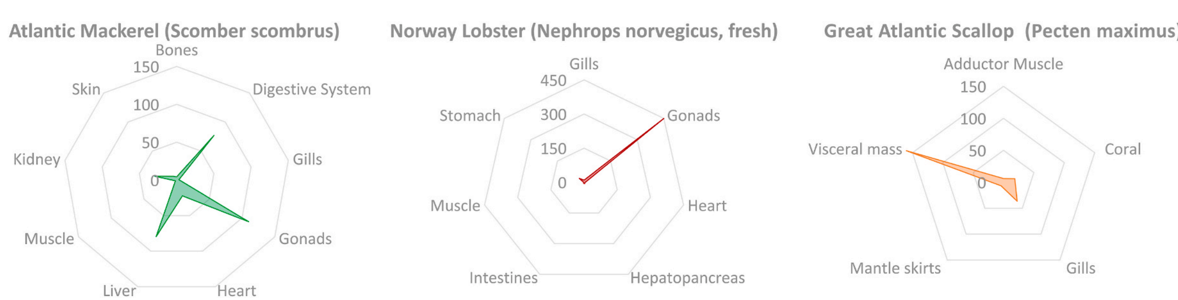

Fig. 3. Activity concentration of ^{210}Po detected in different organs and tissues in three types of seafood (Bq/kg). All activity concentrations refer to fresh weight. Left: Atlantic mackerel (*Scomber scombrus*). Middle: Norway lobster (Nephrops Norvegicus). Right: Great Atlantic scallop (*Pecten maximus*).

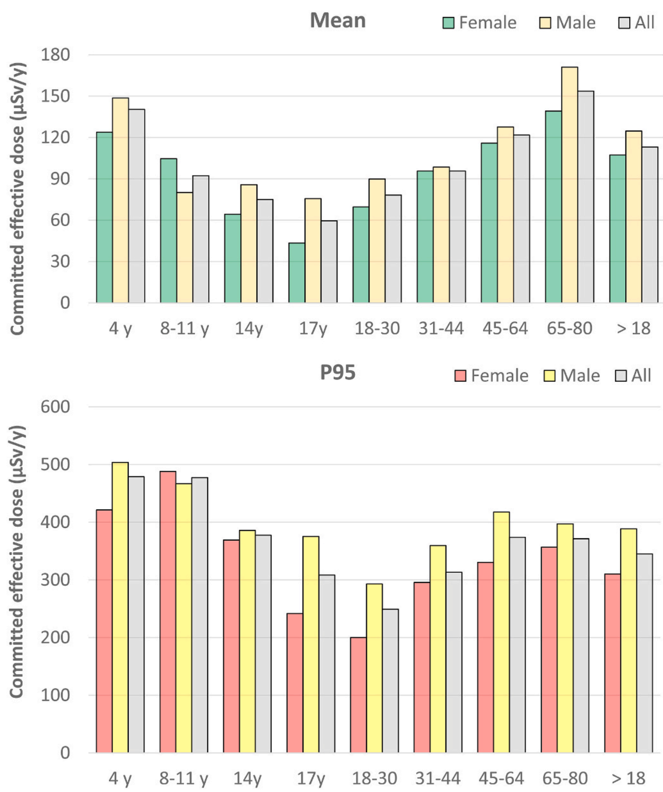

Fig. 4. Committed effective dose from seafood consumption during one year by age and gender. Top: Values based on average values of seafood consumption. Bottom: Values based on 95th percentile of seafood consumption.

crustacean) and Great Atlantic scallop (*Pecten maximus* mollusk), while Fig. 3 summarizes the results. These species represent different trophic levels in the marine food web. Great Atlantic scallop is a primary consumer and suspension feeder (phytoplankton, microscopic algae, bacteria, dead organic matter and/or small organisms out of the water column) (Sealifebase, 2022). Norway lobster is an omnivorous crustacean feeding on other crustaceans, molluscs, polychaete worms, and/or carrion (Institute of Marine Research (Norway), 2022). Whereas Atlantic mackerel is a pelagic fish with a diet rich on zooplankton and small fish (HELCOM, 2013).

The biokinetic distribution of ^{210}Po in the marine biota analyzed was not uniformly distributed. The activity concentration of ^{210}Po ranged as follow: i) Atlantic mackerel between 1.9 ± 0.1 $Bq/kg_{f.w.}$ (muscle) and 110 ± 11 $Bq/kg_{f.w.}$ (gonads) with an average value of 37 $Bq/kg_{f.w.}$; ii) Norway lobster from 2.1 ± 0.1 Bq/kg (muscle) to 450 ± 40 $Bq/kg_{f.w.}$ (gonads), with a mean value of 72 $Bq/kg_{f.w.}$ and iii) Great Atlantic scallop 7 ± 1 Bq/kg (adductor muscle) – 160 ± 18 $Bq/kg_{f.w.}$ (visceral mass), with an average value of 45 $Bq/kg_{f.w.}$. In the three species, high ^{210}Po concentrations were detected in organs and tissues from the digestive system and gonads, while low ^{210}Po content was measured in muscle tissues (see Fig. 3). For instance, the activity concentration of ^{210}Po in Atlantic Mackerel was 40 times higher in the digestive system than in the muscle whereas for Great Atlantic Scallop the ^{210}Po in visceral mass was 27 times higher than in adductor muscle; however, Norway Lobster ^{210}Po concentration was approximately 12 times higher in stomach than muscle tissues. The results support that the intake of ^{210}Po in the marine food web is via biota diet and the bioaccumulation is influenced by their metabolism and different biological processes (Bustamante et al., 2002; Carvalho, 2011; Hansen et al., 2022; IAEA, 2017). In addition, the high concentration of ^{210}Po in the gonads is probably related to the accumulation of protein in the egg and the affinity of polonium to proteins (IAEA, 2017). However, further investigations are needed to identify the biological processes controlling the bioaccumulation of ^{210}Po in gonads and the radiological impact on the genetic material, also mentioned elsewhere (IAEA, 2017). On the other hand, ^{210}Po bioaccumulation in the marine environment is a good indicator of the trophic level position of the biota in the food web (Carvalho, 2011), being lower at upper trophic levels such as Atlantic mackerel than primary consumers such as Great Atlantic scallop (see Table 3 and compare ^{210}Po levels in muscle tissues).

3.3. Committed effective dose

Fig. 4 shows the results of the committed effective dose from seafood consumption during one year from different age groups, and also specified for females and males. Data is given both for mean and P95 annual seafood consumption in Sweden. The average committed effective dose for Swedish population ranged from 60 µSv to 154 µSv, with an average value of 103 ± 31 µSv. The results highlight that committed effective dose higher than 120 µSv was found in the youngest population group (4 years old) because of the high dose coefficient (3.7 times higher than the ^{210}Po dose coefficient for adults) and persons older than 45 years old, due to the high consumption of seafood. The minimum dose was measured in the population group of 17 years old, due to the lowest consumption of seafood. The committed effective dose was controlled by fish consumption below 14 years old, whereas the major contribution to the committed effective dose came from shellfish consumption in people older than 14 years old. The radiological impact of ^{210}Po in males was slightly higher than females, except for the population group of 8–11 years old, where the consumption of seafood was higher for women.

Considering the 95th percentile (P95), the committed effective dose could raise between 249 µSv and 479 µSv, with an average value of 366 µSv, reaching the highest values on population groups below 11 years old (>400 µSv/y). UNSCEAR reported a worldwide exposure to natural radiation from uranium and thorium progeny ranging between 120 and 240 µSv/y (UNSCEAR, 2000). Therefore, in Sweden only the intake of ^{210}Po by seafood reflects the worldwide exposure to natural radiation, highlighting the importance of the monitoring of seafood to control the radiological impact of ^{210}Po on human food chain.

4. Conclusion

This study provided valuable data for better understanding of the radiological impact of ^{210}Po on human food chain to stakeholders, scientific community as well as consumers. The baseline data available in Sweden regarding radioactivity levels on seafood consumed was extended allowing to carry out a more realistic dose assessment. The highest activity concentration of ^{210}Po was detected in Japanese carpet shell (*Ruditapes philippinarum, fresh samples*), blue mussel (*Mytilus edulis, fresh, frozen and canned samples*), oyster (*Crassostrea gigas, fresh samples*), northern prawn (*Pandalus borealis, boiled sample*), European anchovy (*Engraulis encrasicolus, canned*) and skipjack tuna (*Katsuwonus pelamis, canned samples*). Therefore, the consumption of suspension feeders, zooplankton feeders and tuna should be reduced to decrease the intake of ^{210}Po by seafood consumption. In addition, in order to minimize the committed effective dose received by seafood consumption, tissues and organs from the digestive system and gonads should be removed before consumption due to the high levels of ^{210}Po detected. The average committed effective dose for Swedish population ranged from 60 µSv to 154 µSv, with an average value of 103 ± 31 µSv. Moreover, the dose assessment showed that for Swedish population with higher consumption of seafood the committed effective dose could reach values above 400 µSv/y. Therefore, the monitoring of ^{210}Po in seafood is necessary to minimize the radiological impact in Swedish population, in particular, below 4 years old and above 45 years old considering that they were the populations groups with higher values of the committed effective dose.

CRediT authorship contribution statement

F. Piñero-García: Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft, Funding acquisition. **R. Thomas:** Investigation, Resources, Writing – review & editing. **J. Mantero:** Investigation, Writing – review & editing. **E. Forssell-Aronsson:** Resources, Writing – review & editing, Supervision, Funding acquisition, Supervision. **M. Isaksson:** Resources, Writing – review & editing, Supervision, Funding acquisition, Supervision.

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

No data was used for the research described in the article.

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