Activity levels of ^{210}Po in marine organism consumed in south of Spain

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Abstract

The distribution and behaviour of Polonium (^{210}Po) in marine organisms has been and is now a subject of great interest because of their relatively high concentrations in comparison with those in terrestrial organisms and its relatively high weight in the doses received by humans via ingestion. However, depending of the specimen analysed and its relative position along the marine food chain, the ^{210}Po activity levels can be quite different, even in several orders of magnitude. ^{210}Po is primarily absorbed from water and concentrated by phyto- and microzooplankton, and then is transferred to the next trophic level along the marine food chains.

In this work, the contribution of ^{210}Po to the committed effective dose via ingestion received by the Spanish population have been evaluated, by determining the ^{210}Po activity concentrations in an ample set of samples which can be considered representatives of the different marine organisms consumed in Spain. The obtained results show a quite high variability, preventing the possibility to fix a representative value for the ^{210}Po contribution to the ingestion doses received by the Spanish population, but in general these values, due exclusively to ^{210}Po, are higher than the average value assigned by UNSCEAR to the annual committed effective dose received by the worldwide population due to the ingestion of natural and anthropogenic radionuclides.

Introduction

Polonium-210 is a natural occurring radionuclide, belonging to the Uranium series, which is present in minute amounts in the different environmental compartments and that through its route along the trophic chain can finish incorporated in the human body via ingestion of waters and/or food. This radionuclide is highly radiotoxic, with the highest value between the natural radionuclides of the committed effective dose per unit intake via ingestion, and it is present in relatively high concentrations in the marine biota due to its enhanced bioaccumulation and its strong affinity for binding with certain internal tissues. Consequently, ^{210}Po it is an important contributor to the radiation dose received by the marine organisms as well as by the humans consuming seafood.

The high radiotoxicity of ^{210}Po is mainly due to the kind of emissions associated to this radionuclide (alpha particles) and, on the other hand, due to its behavior once it has been incorporated to the human body. According to the ICRP model (ICRP, 1992), for adults, 10% of the inhaled and 50% of ingested ^{210}Po enter the circulatory system while the remaining fraction stays at the gastro-intestinal system for 24-36 hours before being removed by the organism. The absorbed ^{210}Po tends to be accumulated in liver (30%), kidney (10%), spleen (7%) and the bone marrow (10%).
The Spanish population has the seafood as an important component in their diet. Higher committed effective doses via ingestion can be then expected in the Spanish population in relation with other European groups where the culture to include fish in their diet is not so much introduced, due to the higher intake of ^{210}Po associated to this food component. To confirm this fact, we have first estimated the contribution of this radionuclide to the annual committed effective dose by ingestion received by the Spanish population, by determining the ^{210}Po activity concentrations in an ample set of samples which can be considered representatives of the consumed in Spain.

To confirm the key role that the seafood can play for the explanation of the ingestion doses due to ^{210}Po, the levels of this radionuclide in the edible parts of a great variety of marine organisms were determined, with the peculiarity that the analyses were performed after their cooking, in order to check if cooking can alter the ^{210}Po content in the seafood and to refine the dose estimates to human consumers. With this end we have analyzed the ^{210}Po content in the edible parts of several seafood products bought in commercial markets of our town and cooked following the more common recipes in our country. The obtained results will be discussed and evaluated in this work.

Materials and Methods

Samples

The activity concentrations of ^{210}Po were determined in a group of 20 seafood samples purchased in different local markets in Seville (Spain). Also U-isotope determinations were performed in the same samples for comparison purposes. This group of samples was split up in three different subgroups: a) fishes, b) molluscs-crustaceous and c) fish canned preserves.

The fish species samples analyzed were: mackerel (*Scomberscombrus*), dabs (*Dicologoglossacuneata*), atlantic sardines (*Sardinapilchardus*), anchovies (*Engraulisencrasicolus*), europeangiltheads (*Sparusaurata*), hakes considering muscle and spawns (*Merlucciusmerluccius*), and tuna (*Thunnusobesus*), while the molluscs-crustaceous analyzed were: mussels (*Mytilusedulis*), purple dye (*Murex brandaris*), winkles (*Littorinalittorea*), clams (*Donaxtrunculus*),baby clams (*Chameleagallina*), white shrimps (*parapenaeuslongirostris*), king prawns (*Hymenopaenaeusspp*) and cuttlefish (*Sepia betheloti*). Finally, and regarding preserves the following species were analyzed; cockles (*Cerastodermaedule*), anchovies (*Engraulisencrasicolus*) and baby clams (*Chameleagallina*).

Most of these fish species were cooked in the usual way they are consumed for population focusing the posterior radionuclide analyses only in edible parts, with the exception of the preserves which were analyzed directly as they were taken from the can. After cooking, every sample was dried, milled and homogenized before applying a radiochemical procedure for U and Po isotopes isolation and determination.

Radiometric technique

The radiochemical procedure applied for ^{210}Po and U-isotopes determination in the seafood samples, after their pretreatment and after the addition of radiochemical-yield tracers (^{209}Po and ^{232}U respectively), had three main steps: a) a wet digestion process, b) a separation process to isolate Po and U from interfering elements, and c) a source preparation process for alpha measurement.

The digestion process was carried out using a Multiwave 3000 Anton Paar microwave system. This device is equipped with a rotor mechanism with eight XF100 liners (independently guarded) that can work under controlled pressure up to 60 bar and temperatures up to 260 ºC. The liners are sealed in order to avoid any leak of gases during the digestion process, and particularly avoiding Po losses even if the evaporation temperature of this element is exceeded. The liners are made of Teflon and they are inlayed in vessel jacked ceramics making up a stiff reaction cell. The digestion process was performed following the protocol recommended by the manufacturer of the microwave.

The outgoing solution of the digestion process is submitted then to a process of U/Po separation using a well-established liquid solvent extraction with TBP (Holm and Fukai, 1977). Finally, from the resulting two independent solutions containing the U or the Po isolated, the needed thin sources for alpha-spectrometric measurements were performed: in the case of Po applying a self-deposition method onto copper planchets (Flynn, 1968), and in the case of U applying an electrodeposition method onto stainless-steel planchets (Hallstadius, 1984).

The measurement of both U and Po planchets were carried out in a AlphaAnalyst spectrometric (Canberra) system formed by eight separate chambers, each one equipped with a silicon detector (PIPS type) Model A450-18AM, and devoted exclusively to one of the mentioned radionuclides to avoid cross-contamination and for a better background control.

To assess the annual committed effective dose due to ingestion of a particular radioelement associated to each particular sample type, we have applied the following equation:

$$D_E = A \cdot F_C \cdot C$$

where “DE” is the annual committed effective dose (Sv/year) via ingestion of a given marine specie due to the radionuclide under consideration. “A” is the activity concentration of this radionuclide measured in the marine specie considered (Bq / kg wet weight), Fc is the corresponding committed effective dose per unit activity taken by ingestion (Sv/Bq), and C is the amount of the marine specie consumed per person and year, expressed as kg wet weight per year.

The value of Fc is dependent on the radionuclide considered and although it is also depending of the age of the population considered (ICRP, 1992), in this study the annual committed effective doses by ingestion have been determined only for adults. In this case, the values of Fc adopted in this work have been Fc (^{210}Po) = 1.2 10^{-6}Sv/Bq and Fc (^{238}U) = 4.5 10^{-8}Sv/Bq, following ICRP recommendations (ICRP, 1992).

The value of C for each marine specie was obtained from published Spanish statistics concerning food consumption (Ministerio Medio Ambiente, 2006), with the exception of three species, where a estimated consumption of 1 kg per year and person was adopted because no data were available.

Results and Discussion

The activity concentrations of ^{210}Po in the samples representatives of the marine organims consumed by the population of Seville are displayed in Table 1. It is possible to observe that the obtained values are quite variable with differences in some cases of near three orders of magnitude. Also in Table 1 are shown the ^{238}U activity concentrations determined in the same samples, being obtained relatively more uniform values over the time, but clearly lower than the obtained ones for ^{210}Po.

The ^{210}Po activity concentrations (expressed in wet weight) are, in most cases, one or several orders of magnitude higher than those determined for another radionuclide belonging to the same natural radioactive series as ^{238}U.

	^{210}Po (Bq/kg w.w.)	^{238}U (Bq/kg w.w.)
Merlucciusmerluccius (muscle)	2.4±0.7	<0.04
Sardina pilchardus	40±13	<0.07
Engraulisencrasicolus	140±37	<0.19
Thunnusobesus	3.4±1.3	<0.03
Scomberscombrus	17±7	0.03±0.01
Sparusaurata	0.15±0.04	NM
Dicoglosacuneata	28±7	0.03±0.01
Merlucciusmerluccius (roe)	11±3	0.04±0.01
Chamelea gallina	43±12	0.70±0.20
Mitylusedulis 1	84±23	NM
Mitylusedulis 2	115±3	NM
Sepia bethelothi	0.09±0.02	<0.02
Paranaeuslongirostris	21.0±0.5	<0.04
Hymenopaenaeusspp.	0.4±0.1	<0.02
Donaxtrunculus	64±16	0.43±0.11
Bolinusbrandaris	16±4	0.43±0.11
Litorinalittorea	5±1	0.72±0.17
Cerastodermaedule	27±6	0.71±0.15
Engraulisencrasicolus	1.3±0.5	0.07±0.3
Chamelea gallina	10.1±0.1	NM

Table 1. *Activity concentrations of ^{210}Po and ^{238}U (Bq/kg w.w.).*

It is noteworthy to remember that the measurements of ^{210}Po were made on seafood samples that had previously experienced the most characteristic cooking process applied in our geographical area (boiling of shellfish, grilled white fish, etc) since the objective is to perform a dosimetric evaluation as realistic as possible by taking into account the possible losses or redistribution of ^{210}Po between different parts or organs of the analyzed specie in the process of preparation for consumption. The Po activity concentrations obtained were particularly high in some species such as anchovies (140 Bq/kg w.w.), sardines (40 Bq/Kg w.w.), mussels (80-100 Bq/kg w.w.) and clams (60 Bq/kg w.w.).

In fact, and assuming an average value of 1 mBq/L of ^{210}Po in the Atlantic seawater, where the great majority of marine species were collected (Bolivar et al., 2000), concentration factors for ^{210}Po of 10^4- 10^5 can be assessed, indicating the high bioaccumulative behavior of this radionuclide along the marine food chain. The ^{238}U activity concentrations do not overpass, on the contrary, in any case the level of 1 Bq/kg wet weight (Table 1), with concentration factor (CF) several orders of magnitude lower than the found ones for ^{210}Po. All the data shows the existence of a pronounced radioactive fractionation in the uranium series and indicates how wrong can be the simplification of assuming in the marine species secular equilibrium in the uranium series.

The results obtained in this work for ^{210}Po are, on the other hand, of the same order of magnitude to those found in the bibliography (Carvalho, 2011) (Cherry et al., 1994) (Connan et al., 2001) (Kannan et al., 2001) (Pietrzak-Flis et al., 1997) (Alonso-Hernandez et al., 2002) (Dahlgaard, 1996) (Heyraud and Cherry 1979) (Cherry and Heyraud, 1981) (McDonald et al., 1991)(Strok and Smodis, 2011) for the same marine species, although there is a general trend to observe some lower values in this work. This deviation could be due to the previous cooking procedure applied over the samples analyzed in this work, which is not the case in the determinations used from comparison and taken from the bibliography, which corresponds in each specie to edible fresh samples.

The assessment of annual committed effective doses for adults due to ^{210}Po and associated with the ingestion of the different marine species is shown in Figure 1. In this Figure it can be observed that the average annual intake of some of the species, leads to dose values that can be in the order, or exceeds, hundred microsieverts per year (sardines, anchovies, mussels, clams). Then, it can be followed that population having a varied diet rich in marine products could receive a dose due exclusively to ^{210}Po ingestion of seafood in the order of the mSv/year, by simply adding the dose contributions due to different species. This last value can be in a first instance evaluated as quite high, but can be put in context if ,having in consideration the determinations performed in this work, we indicate that simply a consumption of 1 kg of mussels (100 Bq/kg w.w. of ^{210}Po) imply a committed effective dose of 120 microsieverts.

The value of 1 mSv/year is close to that used generically as a global average dose received by people from all natural sources of radiation (2.5 mSv /year), and clearly higher that the average global dose value associated to the ingestion route.

All these facts highlights the importance of the route shown in this study related to the total dose received due to natural sources by the Spanish population. And, additionally, indicates that the above mentioned global average of 2.5mSv/year could underestimate the value of the dose received from all natural sources of radiation that should be associated with the population of Spain.

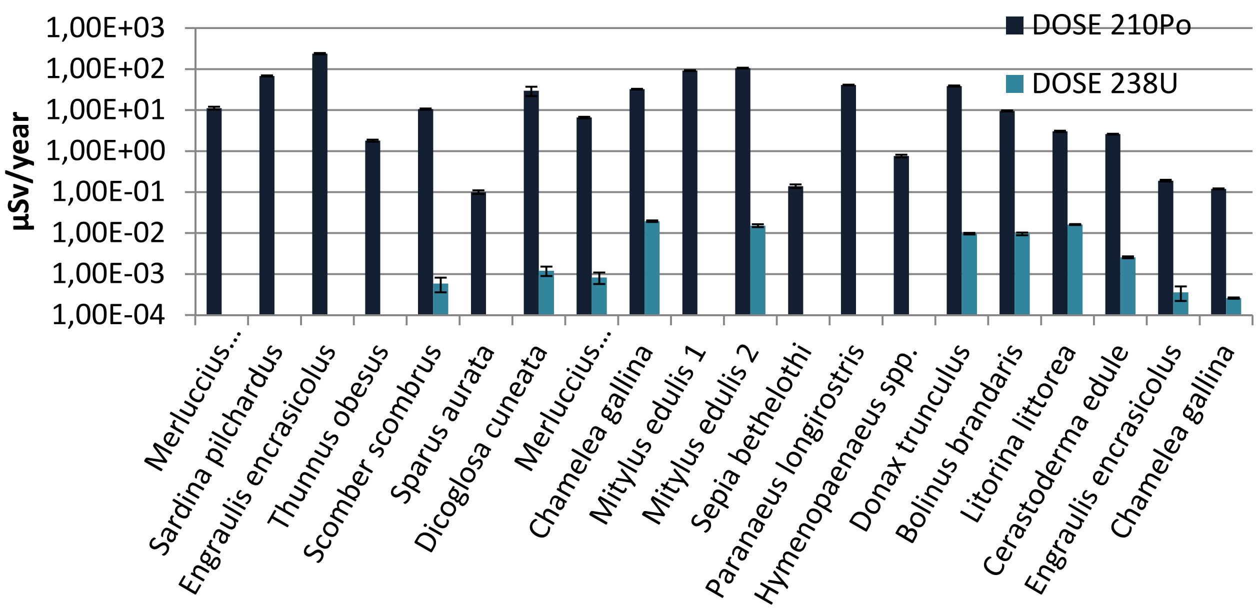

Figure 1.*Doses of* ^{210}Po *and* ^{238}U *(µSv/year)*

To put more in context the value of 1 mSv/year obtained for the annual committed effective dose due to ^{210}Po and associated to the ingestion of seafood by the Spanish population, is interesting to mention that this value is equal to the reference level indicated by the IAEA for the additional occupational doses that could be received in NORM industries without the adoption of countermeasures (IAEA 2006). It is then not surprising to find some industries where some countermeasures are taken in order to decrease the additional dose due to natural radionuclides received by the workers, while at the same time comparable doses can be received by them without any restriction due to their consumption habits.

Finally, and with comparison purposes, Figure 1 also shows the annual committed effective doses for adults due to ^{238}U and associated with the ingestion of the different marine species analyzed in this work. Due to the low activity concentrations found for this radionuclide in the analyzed samples and the low Fc value associated to this radionuclide, the annual committed doses are negligible in comparison with the determined ones for ^{210}Po (in general a factor of 10^4 to 10^5 lower), and in all the cases lower than 10^{-2}µsv/year. These data clearly shows the extremely different role that radionuclides belonging to the same natural radioactive series play along the marine food chain and in the assessment of the ingestion doses received by the population which have the seafood as an important component of their diet.

Conclusions

On the basis of the ^{210}Po determinations performed in composite samples representing the edible parts of an ample set of cooked seafood samples, purchased in the local markets, the annual committed effective dose received by Spanish population due to ^{210}Po and associated to the consumption of these products have been realistically assessed.

Having in consideration the well established culture in Spain for heavy consumption of seafood products, the mentioned annual committed effective dose due exclusively to ^{210}Po can even reach the value of 1 mSv/year, constituting a considerable fraction of the average annual dose received by the Spanish population due to all sources of natural radiation.

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