Evaluation of the Amount of ^{210}Po Ingested by the Spanish Population and its Relation to their Diet Habits

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Abstract

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 diet 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. Knowing the diet habits of the Spanish population and the ^{210}Po bioaccumulative behavior in the marine trophic chain, which implies enhanced concentrations of ^{210}Po in the sea food in comparison with the obtained ones in other components of the diet, the variable and generally higher ingestion doses due to ^{210}Po received by the Spanish population can be associated to the variable and rich consumption of marine products. The Spanish population has the seafood as an essential component of his diet.

Although the ^{210}Po levels in the edible parts of a great variety of marine organisms can be found in the literature, in general the great majority of these determinations correspond to raw edible products. But little is known about the effects of cooking on the ^{210}Po content of seafood which are normally cooked for human consumption. Then it is important to check if cooking can alter the ^{210}Po content in the seafood in order to refine the dose estimates to human consumers. Trying to cover this gap, and at the same time trying to confirm the key role of the seafood in the ^{210}Po ingestion doses received by the Spanish population, we have also 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 conclusion obtained is that for the Spanish population heavily consuming seafood products, the annual committed effective dose via ingestion due exclusively to ^{210}Po can even reach the value of 1 mSv/year ,which constitutes a considerable fraction of the average annual dose received by the Spanish population due to all sources of natural radiation.

Key words: Po-210, cooked diets, seafood, ingestion doses,

1.- 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

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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 diet consumed in Spain. During several years, and every three months, a composite sample formed by several cooked foods and drinks, representing the diet consumed by a person in a week, was collected from a typical Spanish restaurant, and its ^{210}Po content determined as a basis for dose estimation.

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 have been compared with the obtained ones in the same products but analyzed as raw materials and will be discussed and evaluated in this work.

2.- Materials and Methods

2.1 Sample description

The activity concentrations of ^{210}Po (and of ^{234}U and ^{238}U with comparison purposes) were determined within a group of 12 composite samples representative of the diet ingested by the population of Sevilla. For the collection of these composite samples, a local restaurant was selected, where once every three months several cooked plates included in its chart were taken in an amount representing the food which can be ingested by a person in a week. The total mass of each composite sample was about 7 kg w.w. and include different types of meat, fish, vegetables, cereals and several beverages included milk.

Once the composite sample is transported to the laboratory, and after the removal of bones, the process of homogenization starts through the mixture and grinding of the collected edible sample, followed by its drying in an oven at 50ºC during one week to be sure that the total dryness is reached. The dried paste obtained is finally grinded, being ready for radionuclide analysis determination

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

The fish species samples analyzed were: mackerel (*Scomber scombrus*), dabs (*Dicologoglossa cuneata*), atlantic sardines (*Sardina pilchardus*), anchovies (*Engraulis encrasicolus*), european giltheads (*Sparus aurata*), hakes considering muscle and spawns (*Merluccius merluccius*), and tuna (*Thunnus obesus*), while the molluscs-crustaceous analyzed were: mussels (*Mytilus edulis*), purple dye (*Murex brandaris*), winkles (*Littorina littorea*), clams (*Donax trunculus*), baby clams (*Chamelea gallina*), white shrimps (*parapenaeus longirostris*), king prawns (*Hymenopaenaeus spp*) and cuttlefish (*Sepia betheloti*). Finally, and regarding preserves the following species were analyzed; cockles (*Cerastoderma edule*), anchovies (*Engraulis encrasicolus*) and baby clams (*Chamelea gallina*).

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

2.2 Methodology

The radiochemical procedure applied for ^{210}Po and U-isotopes determination in the diets and 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.

^{210}Pb was determined in some seafood samples by gamma-ray spectrometry through its 46,5 keV gamma emission using an extended range Germanium coaxial detector (XtRa) of 37,1% relative efficiency, and after applying self-absorption corrections. This system has a 10 centimetres passive shielding of ancient lead (6 Bq/kg of ^{210}Pb) and an active shielding made with an organic scintillation detector (Bicron BC-418) working in coincident mode with the Ge detector: This combination allow to achieves accurate results in environmental gamma measurements due to its low background level and a quite limited presence of ^{210}Pb in the background spectrum.

2.3 Dose assessment

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

The value of F_c 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 F_c adopted in this work have been F_c (^{210}Po) = 1.2 10^{-6} Sv/Bq and F_c (^{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. The value of C for the composite diets was taken as 1 kg w.w per day (365 kg w.w. per year).

3.- Results and Discussion

The activity concentrations of ^{210}Po in the composite samples representatives of the diet 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 two 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.

Composite Diet simple	^{210}Po (mBq/kg w.w)	^{238}U (Bq/kg w.w)	^{210}Po (μSv/y)	^{238}U (μSv/y)
January-March 2008	5110 ± 30	--	2240 ± 13	--
April- June 2008	180 ± 37	24 ± 12	80 ± 13	0.40 ± 0.23
July- September 2008	202 ± 38	25 ± 10	90 ± 16	0.40 ± 0.17
October-December 2008	421 ± 45	--	185 ± 20	--
January-March 2009	235 ± 22	28 ± 14	102 ±10	0.46 ± 0.23
April- June 2009	73 ± 22	29 ± 14	32 ± 10	0.48 ± 0.24
July- September 2009	890 ± 74	3 ± 3	390 ± 30	0.06 ± 0.06
October-December 2009	615 ± 41	15 ± 6	270± 22	0.26 ± 0.10
January-March 2010	893 ± 47	18 ± 7	390 ± 21	0.30 ± 0.11
April- June 2010	760 ± 43	35 ± 12	332 ± 20	0.57 ± 0.20
July- September 2010	2040 ± 100	38 ± 12	890 ± 42	0.62 ± 0.20
October-December 2010	1810 ± 80	16 ± 7	795 ± 35	0.27 ± 0.12

Table 1.- *^{210}Po and ^{238}U activity concentrations in representative diet samples (Seville, Spain) as well as ^{210}Po and ^{238}U contributions to the annual committed dose received by the population ingesting these diets*

The relatively high levels of ^{210}Po in the diet samples in comparison with other radionuclides from the uranium natural series, together with its high radiotoxicity converts to ^{210}Po as a key contributor to the committed effective dose via ingestion received by the population in Seville (Spain). In the Table 1 are indicated the contribution of ^{210}Po and ^{238}U to the mentioned committed doses calculated with basis in the determined activity concentrations. It is possible to observe how the ingestion doses due to ^{210}Po are much more higher that the ingestion doses due to ^{238}U (several orders of magnitude) and are at the same time quite variable, preventing the last fact the possibility to assign a representative average value to these ingestion doses for the population at the South-West of Spain. In spite of this fact, it is important to indicate that the ingestion doses due exclusively to the contribution of ^{210}Po, and determined from the diet samples analyzed, are in the majority of the cases clearly higher that the value assigned by UNSCEAR to the average whole committed effective doses via ingestion and taken as representative for the worldwide population.

Looking for the importance of the sea food in the annual committed doses by ingestion due to ^{210}Po, the activity concentrations of this radionuclide in marine food regularly consumed by the population of Seville were determined, being the obtained results illustrated in Table 2.

		^{210}Po this work (cooked fish)	**^{210}Po Literature (fresh fish)**
FISH	*Merluccius merluccius (muscle)*	2.4±0.7	6.4±0.3 [1]
	Sardina pilchardus	40±13	66±2 [1]
	Engraulis encrasicolus	140±37	158-203 [2]
	Thunnus obesus	3.4±1.3	3.0±0.1 [1]
	Scomber scombrus	17±7	3.5-19 [1]
	Sparus aurata	0.15±0.04	3.1±0.6 [3]
	Dicologlossa cuneata	28±7	---
	Merluccius merluccius (roe)	11±3	52±2 [1]
MOLLUSKS AND CRUSTACEUS	*Chamelea gallina*	43±12	150 [1]
	Mytilus edulis	84±23	80-220 [4]
	Mytilus edulis 2	115±3	80-220 [4]
	Sepia betheloti	0.09±0.02	0.08 [5]
	Paranaeus longirostris	21.0±0.5	17-810 [6]
	Hymenopaenaeus spp.	0.4±0.1	----
	Donax trunculus	64±16	----
	Bolinus brandaris	16±4	-----
	Littorina littorea	5±1	13-399 [7]
PRESERVES	*Cerastoderma edule*	27±6	6.0 [1]
	Engraulis encrasicolus	1.3±0.5	------
	Chamelea gallina	10.1±0.1	------

Table 2. *Activity concentration (Bq/kg wet weight) of ^{210}Po in seafood samples bought at local markets and cooked before being analyzed (uncertainties 1-σ).*
[1] Carvalho, 2011, [2] Cherry et al., 1994 [3] Connan et al., 2007 [4] Dahlgaard, 1996 [5] Heyrraud and Cherry, 1979 [6] Cherry and Heyraud, 1981 [7] McDonald et al., 1991.

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.).

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 (Table 3).

		^{238}U
FISH	*Merluccius merluccius (muscle)*	<0.04
	Sardina pilchardus	<0.07
	Engraulis encrasicolus	<0.19
	Thunnus obesus	<0.03
	Scomber scombrus	0.03±0.01
	Sparus aurata	NM
	Dicologlossa cuneata	0.03±0.01
	Merluccius merluccius (roe)	0.04±0.01
MOLLUSKS AND CRUSTACEUS	*Chamelea gallina*	0.70±0.20
	Mytilus edulis	NM
	Mytilus edulis 2	NM
	Sepia betheloti	<0.017
	Paranaeus longirostris	<0.04
	Hymenopaenaeus spp.	<0.015
	Donax trunculus	0.43±0.11
	Bolinus brandaris	0.43±0.11
	Littorina littorea	0.72±0.17
PRESERVES	*Cerastoderma edule*	0.71±0.15
	Engraulis encrasicolus	0.07±0.03
	Chamelea gallina	NM

Table 3. *Activity concentration (Bq/kg wet weight) of ^{238}U in seafood samples bought at local markets and cooked before being analyzed. (NM= Not Measured, uncertainties 1-σ).*

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 3), 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 radioactive fractionation between the different elements of the uranium series in the seafood samples analyzed has been observed even between the ^{210}Po and its progenitor ^{210}Pb, as it is reflected in the results compiled in Table 4. In a reduced, but significant, number of samples the ^{210}Pb activity concentrations were determined through the application of the gamma-ray spectrometric technique with an extended range germanium detector (XtRa) being obtained activity concentrations for this radionuclide at least one order of magnitude lower that the found ones for ^{210}Po.

Sample	^{210}Pb	^{210}Po	^{210}Po/^{210}Pb actvity ratio
Scomber scombrus	< 2.4	17	>8
Engraulis encrasicolus	3.2±0.8	141	44
Sardina pilchardus	<3.2	39	>12
Mytilus edulis	3.6±0.6	85	23

Table 4 *Activity concentrations of ^{210}Pb and ^{210}Po (in Bq.kg^{-1} wet weight) and activity ratios ^{210}Po/^{210}Pb determined in some of the samples analyzed in this work (uncertainties 1-σ)*

For the pair ^{210}Po-^{210}Pb is then observed a behavior previously described in the open literature (Carvalho, 2011) (Heyraud and Chery, 1979) with a common pattern: ^{210}Po > ^{210}Pb in marine organisms. Taking then in consideration that the ^{210}Pb/^{210}Po activity ratios in seawater are in the range 0.5-1.0 (Fowler, 2011), it is straightforward to conclude that the ^{210}Po has a higher bioaccumulative behavior than ^{210}Pb along the marine trophic chain.

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 (see Table 2, where literature data are also included). 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 2.5

mSv/year could underestimate the value of the dose received from all natural sources of radiation that should be associated with the population of Spain.

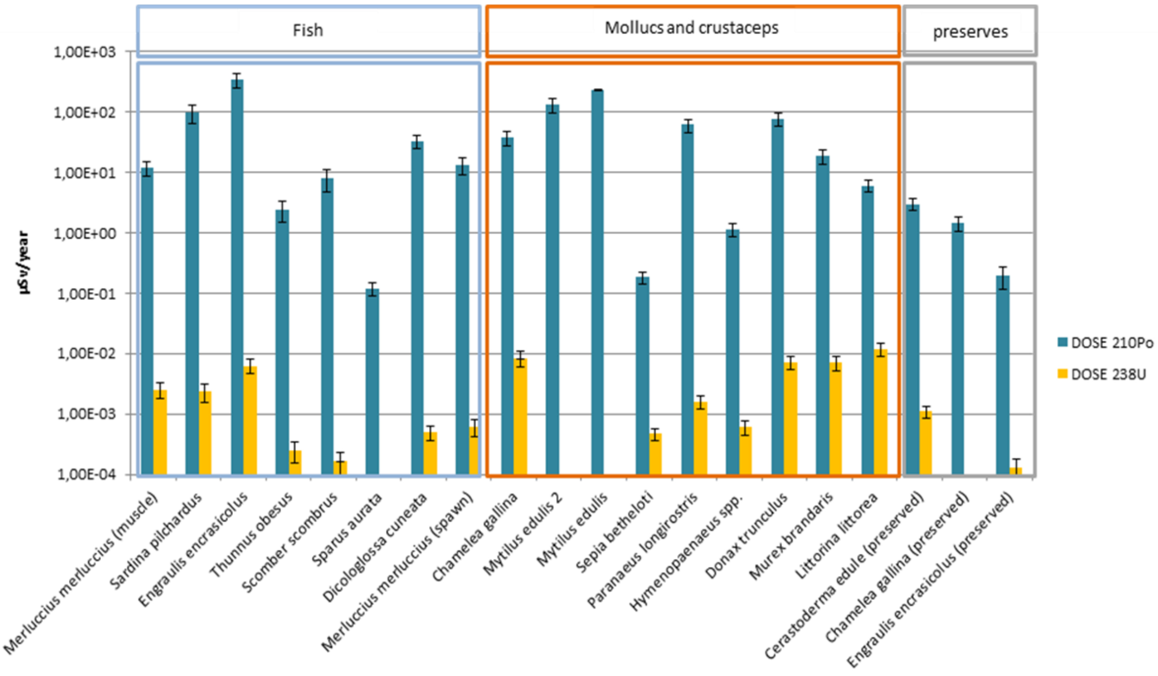

Figure 1. *Committed effective dose in adults (µSv/year) by ingestion assessed in seafood samples due to* ^{210}*Po and* ^{238}*U (error bars 1-σ).*

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 F_c 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 diet of the Spanish population as well as in the edible parts of an ample set of cooked seafood samples, purchased in the local markets of Seville (Spain), the annual committed effective dose received by the 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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