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Research Article

^{210}Po Activity Concentrations in Cooked Marine Food

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Abstract: In this work the ^{210}Po activity concentrations determined in the most common marine species consumed by the population of Southern Spain are shown and discussed. The determinations have been done on aliquots of the raw edible materials and in aliquots previously cooked in the usual way they are ingested. The obtained activity concentrations in the edible parts of the cooked products ranged up to three orders of magnitude with concentration factors for some species as high as 10^4-10^5 L/kg fresh weight, and due to the cooking processes in some cases a considerable variation in the ^{210}Po activity concentrations have been observed in relation with the determined ones in the aliquots not processed. The importance of this study is based in the fact that ^{210}Po is one of the main contributors to the radiation dose received by the population by ingestion, being the ^{210}Po doses by ingestion particularly high when a population has a diet with a high proportion of marine products.

Keywords: ^{210}Po; Marine food; Ingestion doses

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Introduction

^{210}Po is a natural occurring radionuclide, belonging to the uranium series. This radionuclide is present in minute amounts in the different environmental compartments (water, soil, and atmosphere) and through the trophic chain can be finally incorporated in the human body via ingestion of water and/or food. This radionuclide is highly radiotoxic, with the highest value among the natural radionuclides of the committed effective dose per unit intake via ingestion (1.2×10^{-6} Sv/Bq) (IAEA, 2003), 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 tissues (Fowler, 2011). Consequently, ^{210}Po is an important contributor to the natural 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 for adults (ICRP, 1992), a 50% of ingested ^{210}Po enters into the human circulatory system while the remaining fraction stays at the gastro-intestinal system for 24-36 hours before being removed by the organism.

The Spanish population uses seafood as an important component of their diet, whereas this kind of food has a poor proportion in other European diets. So, a higher intake of ^{210}Po and higher committed effective doses via ingestion could be expected in the Spanish population in relation with other European countries.

The ^{210}Po concentrations in the edible parts of a wide variety of marine organisms, including macro-zooplankton and fish, can be found in the literature (Carvalho, 2011; Cherry et al., 1994; Connan et al., 2007; Strok and Smodis, 2011). Usually these determinations correspond to raw edible products; however studies on the effects of cooking on the ^{210}Po content of seafood are very scarce. These studies should be implemented due to the direct consumption of cooked seafood by human beings.

^{210}Po can suffer in some cases volatilization during the cooking process (the ^{210}Po is highly volatile) or migration to cooking wastes which are discharged without be consumed. In other cases, the activity concentration of ^{210}Po can increase due to the loss of weight of the seafood during cooking. The differences between the ^{210}Po activity concentrations found in fishes before and after cooking will be dependent of the fish type under analysis and also it can be dependent of the cooking process applied. For that reason, and in order to have a more realistic information about the ^{210}Po intake by the Spanish population, associated to seafood, the ^{210}Po content in the edible parts of several seafood products, bought in local markets, were performed before and after their cooking following the more common recipes in the region selected: the south of Spain.

Materials and Methods

Sample description and cooking preparations

A total of 9 different kinds of fishes and 2 different kinds of molluscs have been studied in this work **(Table 1)**. In all the fish samples, the edible material collected was separated in two sub-samples: one of them was directly analyzed and the second one was cooked following a procedure described in the coming paragraphs.

The samples of fishes analyzed corresponded to *Scomber scombrus, Salmo solar, Sardine pichardus, Micromesistius poutassou, Mullus barbatus, Xiphias gladius, Gadus morhua, Engraulis encrasicolus, and Solea solea*.

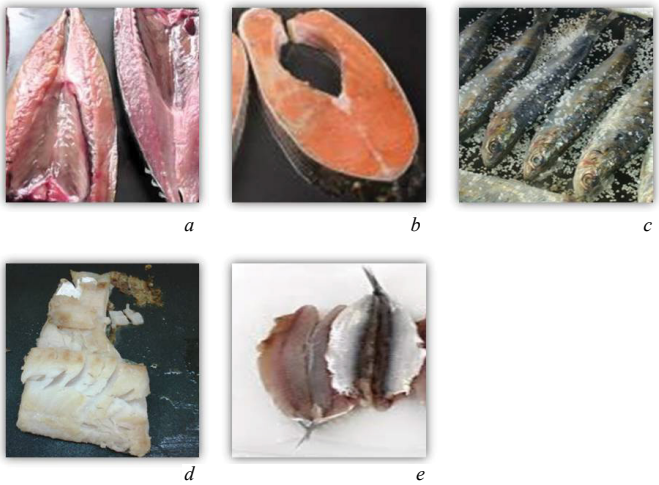
A sample of *Scomber scombrus* (mackerel) was firstly treated. Applied procedure started by removing its head, internal organs and central bone. Then, the rest of this sample was separated in two sub-samples with similar mass **(Figure 1a)**. A sub-sample was cooked by grilling it in a pan at a temperature of 180ºC.

The second sample was salmo solar (salmon), which was bought in a local market. The sample was presented in form of slices **(Figure 1b)**. A number of slices were directly analyzed and the rest of slices were grilled. Skin and bones of sample were removed before analysis in both cases, but after cooking.

Next sample was *Sardine pichardus* (*Sardine*). Here the sample was formed by several units due to the low mass of each sardine. A number of sardines were directly analyzed and a similar number

Table 1: Fishes and molluscs species analysed in this work.

Fishes	
Mackerel	*Scomber scombrus*
Salmon	*Salmo solar*
Sardine	*Sardine pichardus*
Blue whiting	*Micromesistius poutassou*
Red mullet	*Mullus barbatus*
Sword fish	*Xiphias gladius*
Cod	*Gadus morhua*
Anchovy	*Engraulis encrasicolus*
Sole	*Solea solea*
Mollusks	
Clam	*Camelea gallina*
Mussel	*Mitylus edulis*



a *b* *c* *d* *e*

Figure 1: Some of the seafood samples analysed in this work.

of sardines were cooked by grilling. Their heads, internal organs and bones were removed before analyses in both cases, but after cooking. To avoid the pasting during the cooking process, salt was previously added **(Figure 1c)**. The same procedure was applied to other fishes of other species: micromesistius poutassou (blue whiting), and mullus barbatus (red mullet), adding some oil onto the pan to avoid the pasting of the cooked product.

Other analyzed specie was *Xiphias gladius* (Sword fish). The sample was bought in a local market and was presented as fillets of a same fish. A cooked fillet was prepared in an oven with some olive oil, onions and species. The maximum temperature reached in the oven was lower than 200ºC.

Two fresh pieces of *Gadus morhua* (cod) were also bought in the local markets, being one of them cooked by grilling **(Figure 1d)**. Some ml of olive oil was added onto a pan in order to avoid the pasting of the product.

Due to low mass of *Engraulis encrasicolus* (anchovy), a few units were considered in the same sample. These units were cleaned by removing their heads, internal organs and bones **(Figure 1e)**. Each unit was divided in two parts. A part of every unit was used to form two composite samples representative of the whole material. One of these composite samples was cooked by frying it with boiling sunflower oil.

Finally, in the case of the *Solea solea* (sole) specimens, and due to the difficulties for the removal of the skin and the central bones in the raw material, each unit was not divided in two parts. Instead some units were used for the determinations in the raw material and other units were cooked by grilling.

Concerning the molluscs, they were *Camelea gallina* (clam) and *Mitylus edulis* (mussel). Firstly, all the units were immersed in salty water for the removal of the sand that could accumulate. Two sub-samples were separated from the main sample. A sub-sample was vapor boiled, while the raw representative sub-sample was obtained by introducing some units in water with lemon juice.

^{210}Po determination methodology

All the sub-samples (directly analyzed and cooked) were dried, milled and homogenized before applying a radiochemical procedure for Po isolation and determination. The radiochemical procedure, after the addition of a known amount of ^{209}Po as radiochemical-yield tracer, had three main steps: a) a wet digestion process, b) a separation process to isolate Po from interfering elements, and c) self-deposition of polonium to obtain a radioactive source, containing ^{210}Po and ^{209}Po, for alpha spectrometry.

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 260ºC. The liners were 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 was exceeded. The liners were made by Teflon and they were inside a ceramic vessel making up a stiff reaction cell.

The digestion process was performed following a protocol recommended by the manufacturer of the microwave, and the outgoing solution of the digestion process was submitted then to a process of Po separation using a well-established liquid solvent extraction procedure (Holm and Fukai, 1977). Finally, from the resulting solution containing the Po isolated, a thin source for alpha-spectrometric measurement was prepared by applying a self-deposition method onto copper planchets (Flynn, 1968).

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

The software used in the alpha spectra analysis was Genie 2000 from Canberra.

Validation tests

With validation purposes, we have measured duplicate aliquots of two IAEA reference materials. The IAEA-414 (reference date in 1997) is a fish sample from the Atlantic Ocean and the IAEA-437 (reference date in 2003) consists of Mediterranean mussels. Both samples are certified in ^{210}Pb (and we can assume as well that they are certified in ^{210}Po due to the existence of secular equilibrium).

The results obtained for the Po activity concentrations in the reference samples were evaluated by determining the z-score parameter, which is calculated following IAEA documentation (IAEA, 2008). If the absolute value of the z-score is lower than 2, the reported value will be considered as satisfactory; it would be questionable if it is between 2 and 3 and unsatisfactory if it is higher than 3.

Results

Activity concentrations obtained in the performance of the validation exercises are compiled in **Table 2**. Considering the z-score values obtained in both exercises that in all cases are lower than 2, it can be indicated that the methodology employed in this work for ^{210}Po determination is quite satisfactory.

Supporting also the reliability of the results shown in this paper, we can indicate that an average value of 64% was obtained as radiochemical yield of the methodology applied to the whole set of samples that were analyzed in this work (in the range from 50% to 80%) using ^{209}Po as the internal tracer. These radiochemical yields are quite typical to be found when ^{210}Po determinations in biological samples are performed. On the other hand for some of the samples studied, duplicate aliquots were analyzed, being obtained the results compiled in **Table 3**. The obtained results show in all the cases compatibility between the duplicate aliquots, having in consideration the associated uncertainties.

Finally, the whole set of results obtained in the ^{210}Po determinations performed in the fish and mollusc samples analyzed are compiled in **Table 4**. The ^{210}Po activity concentrations determined in the fish raw aliquots were particularly high in some species such as in red mullets, anchovies and sardines (more than

Table 2: Results on the validation tests applied to the whole radiochemical procedure with seafood matrices (1σ-uncertainties, taking in consideration counting uncertainties, and uncertainty in the tracer).

	Matrix	*Target value (Bq·kg^{-1})*	*Reported value (Bq·kg^{-1})*	*Chemical yield (%)*	*Z-score*
IAEA-414	Fish	2.22 ± 0.67	2.59 ± 0.32	71 ± 4	1.67
IAEA-437	Mussels	4.6 ± 0.9	4.1 ± 0.4	64 ± 2	-1.07

Table 3: Results obtained in the duplicate analysis of some of samples studied in this work (1σ -uncertainties, taking in consideration counting uncertainties, and uncertainty in the tracer).

Sample	Sub-sample 1	Sub-sample 2
Mackerel, raw	1.7 ± 0.2	1.7 ± 0.1
Mackerel, grilled	1.8 ± 0.2	1.8 ± 0.1
Sardine, raw	17.8 ± 0.8	18.4 ± 0.7
Sardine, cooked	13.6 ± 0.6	15.7 ± 0.8
Salmon, raw	0.09 ± 0.02	0.06 ± 0.01
Salmon, cooked	<0.05	<0.05

Table 4: ^{210}Po activity concentrations (Bq/kg wet weight) determined in raw and cooked aliquots of the seafood species analysed in this work.

^{210}Po activity concentrations (*Bq·kg^{-1}* w.w)		
Fishes	**Raw**	**Cooked**
Mackerel *(Scomber scombrus)*	1.7 ± 0.2	1.8 ± 0.2
Salmon *(Salmo solar)*	0.09 ± 0.02	<0.05
Sardine *(Sardine pichardus)*	17.8 ± 0.8	13.6 ± 0.6
Blue whiting*(Micromesistius poutassou)*	4.0 ± 0.2	0.76 ± 0.06
Red mullet *(Mullus barbatus)*	32.0 ± 1.1	9.5 ± 0.3
Sword fish *(Xiphias gladius)*	0.40 ± 0.06	0.25 ± 0.02
Cod *(Gadus morhua)*	0.71 ± 0.05	0.80 ± 0.08
Anchovy *(Engraulis encrasicolus)*	21.1 ± 0.7	18.1 ± 0.7
Sole *(Solea solea)*	4.5 ± 0.3	3.6 ± 0.2
Molluscs	**Raw**	**Cooked**
Clam *(Camelea gallina)*	42 ± 2	26 ± 1
Mussel *(Mitylus edulis)*	121 ± 4	59 ± 1

20 Bq.kg^{-1} fresh weight in each case), covering on the other hand a wide range of values. Differences even of two orders of magnitude can be found among the different species analyzed.

Discussion

The results obtained for the unprocessed fish samples in this work for ^{210}Po are of the same order of magnitude as reported by other authors (Carvalho, 2011; Cherry et al., 1994; Connan et al., 2007; Kannan et al., 2011; Pietrzak et al., 1997; Alonso-Hernandez et al., 2002; Cherry and Heyraud, 1981; Dahlgaard, 1996; Heyraud and Cherry, 1979; McDonald et al., 1991; El Samad et al., 2010; Uddin et al., 2016) for the same marine species, although there is a general trend to find slightly lower values in this work. In addition, some correlation can be found between the ^{210}Po levels found and the position of the species analyzed along the trophic chain, with lower values in the ones located in the upper levels.

Assuming an average value of 1 mBq L^{-1} of ^{210}Po in the Atlantic seawater (Bolivar et al., 2000) where the great majority of the fishes were collected, the concentration ratios for ^{210}Po in the unprocessed fishes can reach the values of 10^4-10^5 l/kg fresh weight, indicating the high bioaccumulation behavior of this radionuclide along the marine food chain. The consumption of seafood has a considerable weight in the doses received by the population via ingestion (Diaz-Francés et al., 2014).

The influence of the cooking in the activity concentrations of ^{210}Po in the analyzed fishes can be evaluated on the other hand as quite variable depending of the particular species under consideration. In some cases as for the blue and red mullet species the decrease in the ^{210}Po content is quite considerable after cooking, while in other cases as for cod and mackerel the ^{210}Po levels remain constant after cooking with no lost and/or evaporation. In species with particular high consumption in South of Spain such as sardines and anchovies it can be observed only a small but detectable decrease in the activity concentrations of ^{210}Po due to cooking.

Important variations have been observed on the other hand in the ^{210}Po levels associated to the cooking of the molluscs analyzed. Clearly lower values have been found in the cooked aliquots (particularly in the mussels) indicating the loss of a considerable fraction of this radionuclide either by evaporation or by transfer to the liquid used or formed during cooking by vapor boiling. In some cases, the formed liquids are used independently by the local people in South of Spain for the production of soups and the possible presence in them of ^{210}Po should be considered for a proper dosimetric evaluation of the population by ingestion.

We can then conclude that, depending of the marine species consumed, the estimation of the doses due to the ingestion of ^{210}Po could be overestimated if the estimations are based in the ^{210}Po levels determined in their raw eatable fractions without cooking. In extreme cases of high consumption of marine molluscs this overestimation could be higher than 100%.

Conclusions

The ^{210}Po activity concentrations in different seafood species consumed by the population in the south of Spain, could suffer, depending of the specie analyzed, considerable variations due to the cooking process applied to them. This fact should be taken in consideration for a proper and realistic evaluation of the doses received by the public considered due to ingestion. The estimation of the ingestion doses with basis in the ^{210}Po levels found in uncooked seafood could conduit to no negligible overestimation of the ingestion doses.

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