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^{210}Po in Foodstuffs Consumed by the Population of South-West of Spain

Inmaculada Díaz-Francés[1] and Rafael García-Tenorio[1,2,]*

[1] Applied Nuclear Physics Group, Universidad de Sevilla, Sevilla, Spain
[2] Centro Nacional de Aceleradores (CNA), Sevilla, Spain

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Abstract: The ^{210}Po activity concentrations in a good set of foodstuffs forming part of the representative diet consumed by the population of Seville (Spain) has been determined by alpha-particle spectrometry and the obtained values compared with the published ones for similar products over the world. The results obtained allow confirming the hypothesis reflected in a previous work that the relatively high amount of ^{210}Po ingested by the population of Seville annually associated to their diet, is related with the habit of high seafood consumption.
Keywords: ^{210}Po, foodstuff, ingestion dose.

1 Introduction

In a previous work published in 2016, our research group determined the ^{210}Po activity concentrations in total representative diet samples consumed by the people living in the town of Seville, South-West of Spain [1]. The motivation of this study was based in the following facts:

a)the^{210}Po, due to its high radio toxicity, is the main contributor to the dose received by population due to ingestion, and

b) the previously published ^{210}Po concentrations in diets over the world were quite variable in magnitude, being dependent these concentrations of the diet habits of the population under study.

The conclusion of the study performed was quite clear: the amount of ^{210}Po ingested annually by the population of Seville associated to the diet was quite high in comparison with the ingestion of the same radionuclide determined in other European countries (see Table 1) and, in addition variable over time.

These facts were associated to the comparatively high and variable consumption of seafood products by the Spanish population, because it is well known that the marine biota tends to bio accumulate Po due to the strong affinity of this radionuclide for binding with certain internal tissues [2].The amount of ^{210}Po incorporated by the Sevillian population associated to its diet was only comparable to the levels determined in other countries with a diet particularly enriched in sea-products, as Japan, and clearly higher than the value determined in European countries like Great Britain and Poland where the weight of the seafood in the diet is minor.

Table1. Amount of ^{210}Po (in Bq/y) annually ingested by the population of different countries over the world associated to their diet.

Country	^{210}Po, Bq/year ingested	Ref.
Spain	275	(1)
Japan	250	(3)
Italy	150	(4)
Poland	50	(5)
Great Britain	35	(6)

*Corresponding author e-mail: gtenorio@us.es

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In order to confirm the hypothesis previously indicated for explaining the ^{210}Po diet results in Seville, the authors have determined the ^{210}Po activity concentrations individually in a good number of the foodstuffs forming part of a representative Seville diet.

These determinations have been performed in beverages, cereals and legumes, eggs and milk products, leafy and root vegetables, fruits, meat and fish products collected from local markets. The discussion of the results obtained in this study and the comparison with the obtained ones in the same or similar products over the world form the core of this paper.

2 Experimental Sections

Being the ^{210}Po a pure alpha emitter, its determination has been performed by applying the alpha-particle spectrometric technique. This implies the need the extraction and isolation the ^{210}Po from the product to be analysed.

In this study, representative aliquots of the solid products to be analysed, were first dried at low temperature (to avoid the volatilization of the polonium), spiked with a known amount of ^{209}Po to be used as tracer, and afterwards submitted to a microwave based wet-digestion following the procedure described in [1].

The solution obtained from the digestion is afterwards conditioned in the form HCl 2M and the Po self-deposited over cupper planchets, following the procedure described in [7]. In the case of liquid samples, the preparation method described in [7] was applied as a whole.

The Po planchets were measured using an alpha-particle spectrometric system, Alpha-Analyst from Canberra Co., formed of a total of eight independent chambers working in parallel, each one equipped with a PIPS type silicon detector (450 mm^2 active area).The measurements were performed with a fixed-distance source detector of 1.2 cm, corresponding to a geometric efficiency of around 25 %.

With this efficiency and for the range of radiochemical recoveries obtained after the application of the radiochemical procedure for their isolation and conditioning for the measurements, typical minimum detectable activities in the order of 10^{-1}mBq were reached. The software Genie 2000 (Canberra Co.) was used for the analyses of the alpha spectra.

The procedures of ^{210}Po determination in biological and water samples have been validated in our laboratory through the regular participation in intercomparison exercises.

3 Results and Discussion

The ^{210}Po activity concentrations (expressed in wet weight) determined in a good set of different foodstuffs and collected from local markets, are compiled in Table 2 (beverages and cereals and legumes), Table 3 (eggs and milk products), Table 4 (meat, vegetables and fruits) and Table 5 (seafood).

In the mentioned tables a column indicating the values found in the open literature in the same or similar products collected over the world have been also included.

Table 2. ^{210}Po activity concentrations (mBq/kg ww) in beverages, cereals and legumes

SAMPLE TYPE	THIS STUDY	OTHER STUDIES
BEVERAGES		
Tap Water	0.25 mBq/kg	< 1 mBq/kg (8)
Mineral Water	0.6 – 40 mBq/kg Average 5 mBq/kg	0.4 – 6 mBq/kg (9), 0.1 – 21 mBq/kg (10) 0.4 – 4 mBq/kg (11), 2 – 20 mBq/kg (12)
Wine	Red wine 40 mBq/l	Red wine Italy 70 mBq/l (13) Red wine New Zeeland 13 mBq/l (14)
CEREALS AND LEGUMES		
Bread	130 mBq/kg	40-200 mBq/kg Germany (15) 115 mBq/kg England (6)
Rice	200 mBq/kg	50 mBq/kg (4), 75 mBq/kg (14) 230 mBq/kg (16)
Pasta	40 mBq/kg	30 mBq/kg (4) 50 mBq/kg (14)
Legumes	20 mBq/kg (lentis)	12 mBq/kg , beans (14) 23 mBq/kg, peas and beans (6)

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Table 3. ^{210}Po activity concentrations (mBq/kg ww) in eggs and different milk products

SAMPLE TYPE	THIS STUDY	OTHER STUDIES
EGGS AND MILK PRODUCTS		
Eggs	250 mBq/kg	20 – 230 mBq/kg (4) 500 mBq/kg (5)
Cow milk	8 mBq/l	< 9 mBq/l (14) 13- 45 mBq/l (17) 9 – 17 mBq/l (4) 5 – 10 mBq/l (18)
Milk Products	30 mBq/kg, yoghourt	40 – 60 mBq/kg, cheese (4) 50 mBq/kg, dairy products (6)

Table 4. ^{210}Po activity concentrations (mBq/kg ww) in meat, vegetables and fruits.

SAMPLE TYPE	THIS STUDY	OTHER STUDIES
MEAT PRODUCTS		
Bovine	45 mBq/kg	30 mBq/kg (14), 100 mBq/kg (4)
Pork	200 mBq/kg	180 mBq/kg (14), 100 mBq/kg (5)
Chicken	12 mBq/kg	40 mBq/kg (6)
VEGETABLES AND FRUITS		
Leafy Vegetables	170 mBq/kg spinach , 50 mBq/kg chard 25 mBq/kg broccoli 65 mBq/kg lamb´s lettuce 10 mBq/kg Endive, 45 mBq/kg lettuce	200 – 400 mBq/kg spinach (19) 80 – 200 mBq/kg chard (19) 15 mBq/kg broccoli (14)
Root Vegetables	25 mBq/kg potatoes	11 -16 mBq/kg potatoes (14,6) 20 mBq/kg onions (4), 20 – 60 mBq/kg carrots(16)
Fruits	Orange 30 mBq/kg Apple 55 mBq/kg	55 mBq/kg Apple (5) 20 mBq/kg fresh fruits (6), Orange 20 mBq/kg (14)

Table 5. ^{210}Po activity concentrations (mBq/kg ww) in seafood.

SAMPLE TYPE	THIS STUDY	OTHER STUDIES
SEAFOOD		
Main Fishes and Cephalops		
Hake	2.5 Bq/kg	6-7 Bq/kg (15)
Tunna	3.5 Bq/kg	2 – 3 Bq/kg (15)
Mackerel	1.7 Bq/kg	
Small Fishes consuming plankton		
Anchovies	140 Bq/kg	
Sardines	40 Bq/kg	65 Bq/kg (15)
Crustaceus and Shellfish		
Prawn	20 Bq/kg	25 Bq/kg (15)
Mussels	85 Bq/kg	50 Bq/kg (15)
Clams	40 Bq/kg	

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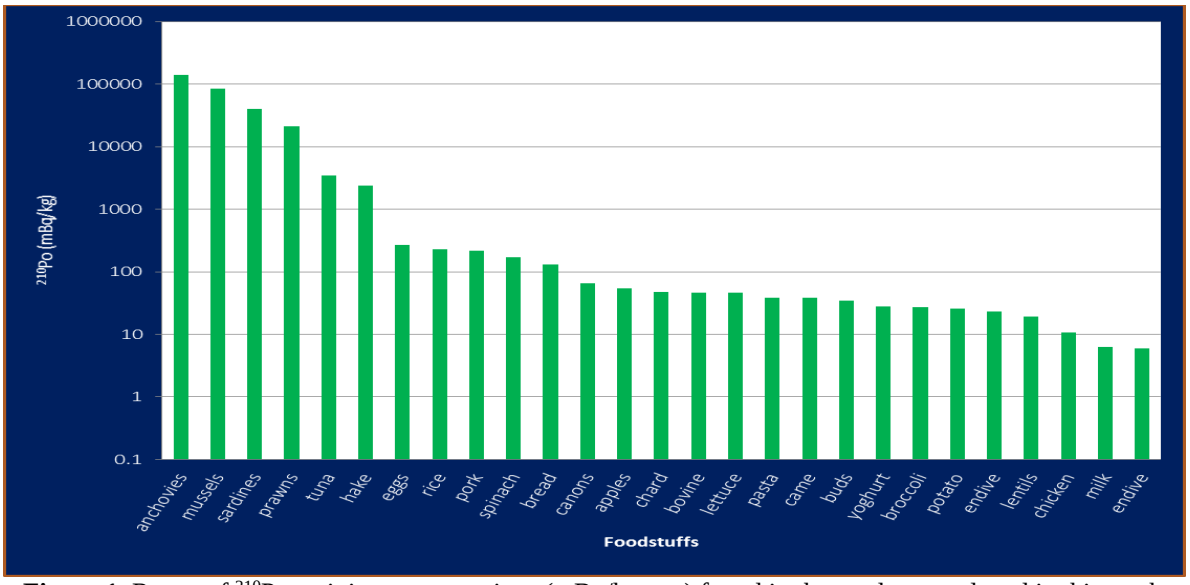

Figure 1. Range of ^{210}Po activity concentrations (mBq/kg ww) found in the products analyzed in this work

The results compiled in the mentioned tables deserve the following general comments:

a) The ^{210}Po activity concentrations determined in the products collected in the local markets of Seville are, in the great majority of cases, similar or comparable to the found ones in other parts of the world. In all the cases, and for the same product, the results determined in this study and the found ones in the open literature are of the same order of magnitude.

b) On the contrary, the obtained results evidence the existence of an enormous range of ^{210}Po activity concentrations between the different foodstuffs. As it can be graphically observed in Figure 1, this range cover even five orders of magnitude, with activity concentrations in the order of mBq/kg (as in the case of tap water) until more than 10^5mBq/kg (in some small fishes consuming plankton).

c) The enormous differences in the 210Po activity concentrations found between the different foodstuffs can explain why are quite variable the amount of 210Po ingested in different countries over the world. Depending of the local diet habits and the major or minor proportion of some foodstuffs in the diet, the amount of ^{210}Po ingested can vary considerably.

d) From the data reflected in Tables 2 to 5, is immediate to conclude that the higher ^{210}Po concentrations are found associated to the seafood

e) are found associated to the seafood products. In all the seafood analyzed, the activity concentration was higher than 10^0Bq/kg. Concentrations higher than 10^{-1}Bq/kg were found in eggs, rice, bread, pork meat, as well as in some leafy vegetables (in this last case, especially in the vegetables with higher capitation surface of the ^{210}Pb-^{210}Po formed in the atmosphere as ^{222}Rn daughters).

f) Concerning the results obtained in the analysis of the seafood products, it is possible to observe a big difference in the levels found between the three groups in which the data have been classified in Table 5. The higher concentrations are found in small fishes that bioaccumulate polonium in association with the plankton that they ingest, as well as in some filter feedings species that accumulates the Po associated initially to the organic and particulate material retained through their filtering processes. The values obtained for main fishes evidence that the concentrations of ^{210}Po diminishes when the products analyzed belongs to higher levels in the marine trophic chain.

g) The products included in tables 2 to 5 are the more representative ones (due to its high consumption) in the diet of the population of Seville. For that reason, all the meat results shown in Table 4

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correspond to farm animals and all the fruits and vegetables results included in Table 4 correspond to cultivated products. Although is known that wild meat generally presents higher ^{210}Po concentrations that farm meat [21], the consumption of wild meat for the population of Seville is anecdotic. Similar case occurs with the consumption of wild mushrooms and forest products such as berries. Quite high values of ^{210}Po can be found in these products [22, 23] but their annual consumption in south-west of Spain is extremely limited.

h) The set of results obtained and the clear differences in the ^{210}Po concentrations between the different components of the diet, clearly explain the high differences observed in the amount of ^{210}Po ingested annually in different countries around the world. A diet enriched in terrestrial products, based in vegetables cereals and meat implies the ingestion of less amount of ^{210}Po that the ingested associated to a diet enriched particularly in marine products.

In fact, classifying all the products analyzed in this work in 8 different generic groups (milk products, meat products and eggs, cereals and legumes, leafy vegetables, root vegetables, fruits, water and beverages. and seafood), and associating to each group an average ^{210}Po activity concentration (mBq/kg ww) (with basis in the experimental results obtained) and an average amount (Kg ww) ingested per year, an estimation of the total amount of ^{210}Po ingested by the population of Seville can be determined, and the relative importance of each group in this total amount can be evaluated. In addition the estimated amount in this way, can be compared with the determined ones in the previous work (1) based in determinations performed in representative total diet samples The indicated estimations are compiled in table 6, and deserves some general comments:

a) The estimated annual amount of ^{210}Po ingested by the population of Seville based in the measurements performed in the individual products (241,5 Bq/y) is in quite good agreement with the value determined with basis in the determination performed in representative total diets (275 Bq/y, see Table 1). This agreement gives confidence about the representativity of the foodstuff products that have been considered in this work in the sevillian diet, and about the approach followed in this work.

b) The extremely high weight of the seafood products in the total amount of ^{210}Po ingested is clearly evidenced. If the seafood group is not considered the total amount of ^{210}Po ingested by the population of Seville does not reach the amount of 50 Bq in clear agremenet with the amounts annually ingested in countries like Great Britain and Poland (see Table 1) where the consumption of seafood products is quite limited.

c) The percentage of ^{210}Po ingested associated to beverages and milk is quite limited and represents a quite small fraction of the ^{210}Po ingested by the population. This contribution is even limited taking into consideration the tendency observed in the last years of an abrupt increase in the consumption of mineral water by the Spanish population. Although some mineral waters present ^{210}Po activity concentrations clearly higher than the found in treated tap waters, their contribution to the total amount of ^{210}Po ingested is small in comparison with the seafood

d) Possible variations in the estimations compiled in Table 6, either in the average ^{210}Po activity concentrations or in the consumed amounts assigned to each foodstuff group does not vary appreciable the total amount of ^{210}Po ingested, in the majority of cases, with only one exception: the variations associated to the seafood group. Only as an example we show in Table 7, a) the variations in the total amounts of ^{210}Po ingested annually by the population of Seville as a function of the amount of seafood consumed, and , in each case, b) the relative percentage of the polonium ingested associated to seafood. The total amount of ^{210}Po ingested can varied from 90 to 290 Bq/y (a factor of 3) when the consumption of seafood changes from 10 to 50 kg per year, and the percentage of ^{210}Po ingested associated to the seafood in relation to the total varies from 55 to 83% in the same interval of seafood consumption.

e) The evaluation performed in the previous paragraphs evidence how sensitive will be the amount of ^{210}Po ingested annually to the amount of seafood consumed by the population of Seville. Different individuals with different habits in the consumption of seafood will ingest quite different amounts of ^{210}Po. This sensitivity is on the contray clearly lower in countries where the seafood play a minor role, being in these case more uniform the amounts of ^{210}Po ingested.

f) This last comment should be taken in consideration in order to understand the difficulties to fix a background value of the ^{210}Po

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associated to the urine as a basis for the fixation of threshold levels in emergency situations where a critical group of population should be screened for possible contamination by ingestion of ^{210}Po. This threshold level is relatively easy to be fixed for population with diets enriched in terrestrial products, but quite difficult

Products, but quite difficult to define, due to its intrinsic variability, for the population with diets enriched in seafood products. A simple extrapolation of threshold levels from one country to another without to take into account the peculiarities of their diets can have as a consequence the adoption of not appropriate decisions.

Table 6. Estimation of the amount of ^{210}Po ingested annually by the population of Seville based in the habit diets of the population and the estimated ^{210}Po activity concentrations associated to each foodstuff group.

Foodstuff	Average activity concentration (mBq/kg ww)	Amount ingested per year kg ww/y	^{210}Po ingested Bq/y
Milk Products	20	100	2
Meat Products, Eggs	100	80	8
Bread, Rice, Pasta, Legumes	70	150	10.5
Leafy vegetables	100	70	7
Root Vegetables	25	80	2
Fruits	50	140	7
Water, Beverages	10	500	5
		Subtotal	**41,5 Bq/y**
Fish	5000	40	200
		Total	**241,5 Bq/y**

Table 7. Variation of the estimated total amount of ^{210}Po ingested by the population of Seville as a function of the amount of seafood ingested and associated percentage of the total ^{210}Po ingested associated to the consumed seafood.

Amount of seafood ingested Kg	^{210}Po Bq/y Total diet	^{210}Po Bq/y seafood	% ^{210}Po ingested due to fish
10	90	50	55.5
20	140	100	71.4
30	190	150	78.9
40	240	200	81.6
50	290	250	82.9

4 Conclusions

In this work, the ^{210}Po activity concentrations in a good set of different foodstuffs forming part of the diet consumed by the population of Seville, Spain, have been determined. The obtained results have allowed; a) to confirm that the total amount of ^{210}Po ingested annually by the citizens of Seville is high in comparison the worldwide average value, and b) to demonstrate that these high values are associated to the important role of seafood in the diet.

In addition, all the poducts analyzed have been assigned to one of the following 8 groups: 1-milk products, 2-meat products and eggs, 3-cereals and legumes, 4-leafy vegetables, 5-root vegetables, 6-fruits, 7-waters and beverages, and 8-seafood. This classification has allowed to estimate the weight of each group in the total amount of ^{210}Po ingested by the population of Sevilla

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