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Analytical Methods

A sequential determination of ^{90}Sr and ^{210}Po in food samples

S. Hurtado-Bermudez [a,*], J.L. Mas [b], M. Villa-Alfageme [c]

[a] Centro de Investigación Tecnología e Innovación, CITIUS, Av. Reina Mercedes 4B, 41012 Sevilla, Spain
[b] Dpto. Física Aplicada I, Escuela Universitaria Politécnica, Universidad de Sevilla, Spain
[c] Dpto. Física Aplicada II, ETSIE, Av. Reina Mercedes 4A, Universidad de Sevilla, 41012 Sevilla, Spain

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ABSTRACT

The latest EU Council Regulation 2016/52/Euratom updates the emergency limits on radionuclides in foods including ^{210}Po and ^{90}Sr, two of the most important radionuclides for radiological dose from the ingestion pathway. A novel and straightforward method has been developed for sequential determination of ^{90}Sr and ^{210}Po in food samples using ultra low-level liquid scintillation counting and alpha-particle spectrometry. For ^{90}Sr analysis, the method makes use of stable strontium as yield tracer, and ^{210}Po is determined through self-deposition using ^{209}Po as a yield tracer. The quantification limit for this method is 25.0 and 2.0 Bq kg^{-1} for ^{90}Sr and ^{210}Po, respectively. The proposed radiochemical separation can be completed within 2 days for a batch of 12 samples. The radiochemical procedure was validated by its application for the measurement of IAEA certified reference materials, and through participation in anational intercomparison exercise. Results are also presented in seafood from the Mediterranean coast. .

1. Introduction

The EU Council Regulation 2016/52/Euratom, and the regulations of other countries (Health Canada., 2000; U.S. Food and Drug Administration, 2004; FSCJ., 2011) and international organizations (United Nations FAO/WHO, 1995), establish a maximum tolerated concentration of radioactivity in food following a radiological emergency.

Amongst all of the radionuclides included in the EU regulation we focus on ^{210}Po and ^{90}Sr. ^{90}Sr is one of the most important radionuclide of anthropogenic origin in the marine environment. Furthermore, ^{210}Po is one of the most characteristic radionuclide of natural origin presents in marine ecosystems.

In the ^{238}U natural decay series, ^{226}Ra disintegrates in a number of successive short-lived daughter nuclides (^{222}Rn, ^{218}Po, ^{214}Pb, ^{214}Bi, ^{214}Po) before ^{210}Pb is formed. ^{222}Rn is a gas that is continually emitted from the earth's surface. The daughter products of ^{222}Rn are associated with particles and washed out of the atmosphere into the sea. In the environment, ^{210}Po ($T_{1/2}$: 138.4 days) is an alpha emitter which is mainly produced from the natural decay of ^{210}Pb ($T_{1/2}$: 22.3 years) through the decay of ^{210}Bi ($T_{1/2}$: 5.01 days) (Matthews, Kim, & Martin, 2007).

Additionally industrial activities can release ^{210}Po in the marine environment. During the EU Marina II project, studies regarding the radiological impact of NORM releases on the European marine systems (comprising gas and oil activities) were carried out (Betti et al., 2004). This study found a widespread distribution of ^{210}Po into the food chain through inhalation, direct uptake or ingestion.

Regardless of the origin of ^{210}Po, its measurement and detection in food is crucial because it is one of the most toxic radionuclides (Guérin & Dai, 2014), which gives rise to health impacts on humans and other organisms because it is an alpha-particle emitter, and contributing to most of the radiation dose received by marine life (UNSCEAR., 2008). Specifically, ^{210}Po is preferably accumulated in protein thus permitting its access to the food chain and increasing levels of ^{210}Po in protein-rich diets including seafood and meat (Watson, 1985).

Many methods have been developed for measuring the radioactivity concentration of ^{210}Po (Matthews et al., 2007). Conventional analytical methods are based on alpha-particle spectrometry and usually involve self-deposition on silver discs. In some sample matrices an extraction and separation step are necessary because interferences may appear from different radionuclides resulting in a massive source deposit or low Po recovery (Matthews et al., 2007).

Regarding ^{90}Sr, it has been considered a key radionuclide in a nuclear accident (i.e. recently Fukushima Daiichi NPP on 2011) because of its high generation rate during nuclear reactions. (Habibi, Boulet, Gleizes, Larivière, & Cote, 2015). Furthermore ^{90}Sr has a long biological and nuclear half-life (nuclear $T_{1/2}$: 28.9 years) becoming a hazardous radionuclide due to its similar

* Corresponding author.
E-mail address: shurtado@us.es (S. Hurtado-Bermudez).

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metabolism than calcium. ^{90}Y, ^{90}Sr daughter nuclide, emits high-energy beta particles increasing the risk of developing bone cancer through its accumulation in bone tissue. This leads to the need of monitoring the ^{90}Sr content in food, and particularly in seafood, and to quantify the absorbed radiological doses through its ingestion.

The Commission Implementing Regulation (EU) 2016/6 of 5 January 2016 set out special restrictions on the import of food from Japan following the accident at the Fukushima nuclear power plant, and Council Regulation (Euratom) 2016/52 of 15 January 2016 established maximum authorized levels of radioactive contamination of food following a radiological emergency. The need to test additional foodstuffs demands the development of new analytical methods in order to quantify the selected radionuclides.

Many methods have been developed for measuring the radioactivity concentration of ^{90}Sr (Matthews et al., 2007; Vajda & Kim, 2010). The most popular methods used for the detection of ^{90}Sr in environmental samples involve liquid scintillation and proportional counter but they are time-consuming methods (Vajda & Kim, 2010).

The purpose of this work is to establish a sequential analytical method for the determination of ^{210}Po and ^{90}Sr radioisotopes in food. The method is based on the extraction chromatography with Sr-resin, followed by strontium precipitation as oxalate and polonium self-deposition, and finally their determination by liquid scintillation counting and alpha-particle spectrometry. The method validation and quality control of the proposed method was carried out using several reference materials.

2. Materials and methods

2.1. Instrumentation

An ultralow background liquid scintillation spectrometer, Quantulus 1220™ (PerkinElmer, Inc.), was used for Liquid Scintillation Counting (LSC). The detector background is reduced by means of combined passive and active shields. The classification of pulses produced by alpha or beta particles is carried out through a pulse shape analysis (PSA) circuit (Villa, Manjón, & García-León, 2003).

An alpha-spectrometry instrument (Alpha Analyst, Canberra) containing twelve independent chambers, each of them with a Passivated Implanted Planar Silicon (PIPS) detector inside, was utilized for the measurement of polonium sources. Each PIPS detector has a good alpha-peak energy resolution of 18 keV (as Full-Width at Half-Maximum FWHM), and an active surface area of 450 mm^2. The polonium sources were located at 1.5 mm from the PIPS detector in order to get high counting efficiency. Alpha spectrum analysis was performed by using Alpha Analyst software (Villa, Hurtado, Manjón, & García-Tenorio, 2007).

2.2. Reagents and analytical solutions

^{90}Sr samples were prepared for LSC through mixing with scintillation cocktail OptiPhase HiSafe III and placing into polyethylene vials (Packard BioScience).

Deionised water (DI water, Millipore) with 18.0 MΩ cm^{-1} resistivity, and Suprapure grade chemicals (HNO_3, H_2O_2, NH_3OH and HCl from Merck) were utilized for solution matrix matching, samples leaching, and chromatographic isolations, when required. Oxalic and ascorbic acid (Panreac, Spain) were analytical grade. Radiochemical yield was determined through analytical grade strontium nitrate $Sr(NO3)_2$ (Sigma-Aldrich). Chromatographic isolation of ^{210}Po and ^{90}Sr were carried out using 2 mL Sr resin columns (TRISKEM, France) placed on a 12-hole polycarbonate vacuum box with an attached pressure valve.

^{210}Po activity and radiochemical yield determinations were performed using a ^{209}Po standard solution (Eckert & Ziegler) of 100 ± 3 Bq g^{-1} (1366-12). The calibration of the LSC system was carried out with a ^{90}Sr standard solution of 107.1 ± 0.4 Bq g^{-1} (MRC-2006-011) obtained from CIEMAT (Spain).

Finally, the energy calibration of the alpha-particle spectrometer was performed with an electroplated alpha standard source of natural U prepared from an aqueous solution of uranyl nitrate hexahydrate ($UO_2(NO_3)_2{\cdot}6H_2O$) (Panreac, Spain). ^{241}Am standard electroplated source from PTB (Germany) containing 93.3 ± 1.9 Bq of (380–911) was used to determine the counting efficiency for each chamber and located at 1.5 mm from the detector. The obtained values range from 0.245 to 0.263 depending of the chamber. The uncertainty of the counting efficiency was less than 3% within a confidence probability of 95%.

2.3. Certified reference materials

The method presented in this work was validated by the analysis of certified reference materials (CRM) provided by the International Atomic Energy Agency (IAEA). The IAEA-437 is a reference material designed for the determination of anthropogenic and natural radionuclides in mussel (*Mytilus galloprovincialis* species) including ^{210}Po (median value of 4.2 Bq kg^{-1}). The IAEA-330 is a certified reference material for radionuclides in spinach including ^{90}Sr (20.1 ± 2.1 Bq kg^{-1}). And finally, the IAEA-414, a mixed fish species from eastern Irish Sea, is a reference material certified for ^{210}Pb (^{210}Po in equilibrium with a median value of 2.1 Bq kg^{-1}) and ^{90}Sr (with a median value of 0.28 Bq kg^{-1}).

2.4. Samples

Seafood samples (sea urchins, mussels and oysters) were taken along the southern Spanish Atlantic coast (Andalusia) and Ballearic Islands in the year 2015. All samples were stored in bags, kept cool using ice and transported to the laboratory. Then samples were placed in filtered seawater for a 24-h period, and afterwards they were cleaned and shucked with a knife in order to separate the soft parts. Each sample was weighed and then dried to constant weight at 60 °C and weighed again. Finally, the samples were homogenized and stored in plastic bags.

2.5. Analytical procedure

The radiochemical method started with sample pre-treatment consisting of digestion with hydrogen peroxide and nitric acid. The next step was the ^{210}Po and ^{90}Sr isolation using the chromatographic Sr columns. Finally, the ^{210}Po and ^{90}Sr sources were prepared in order to measure them with the appropriate technique. The radiochemical procedure is schematized in Fig. 1 and the steps are detailed as follows:

Step 1. *Pre-treatment of samples.* Firstly, if the samples were not previously lyophilized, they were freeze-dried, determining moisture content through weight loss due to drying, and subsequently ground to powder. Then the food samples were easily dissolved taking care of the operating temperature in order to avoid losses due to polonium volatilisation. It has been reported that polonium losses occur leaching biological materials above 100 °C, and more than 90% of the ^{210}Po may be volatilized if the temperature exceeds 300 °C (Martin & Blanchard, 1969). Therefore, a sequential procedure for simultaneous determination of ^{90}Sr and ^{210}Po should avoid previous sample calcination (at 500–600 °C) utilized in most of ^{90}Sr analysis procedures (Vajda & Kim, 2010).

Consequently a dry weight of 5 g from each food sample was put into a Teflon beaker. The high sample capacity of the proposed pre-treatment step outperforms other analytical techniques such

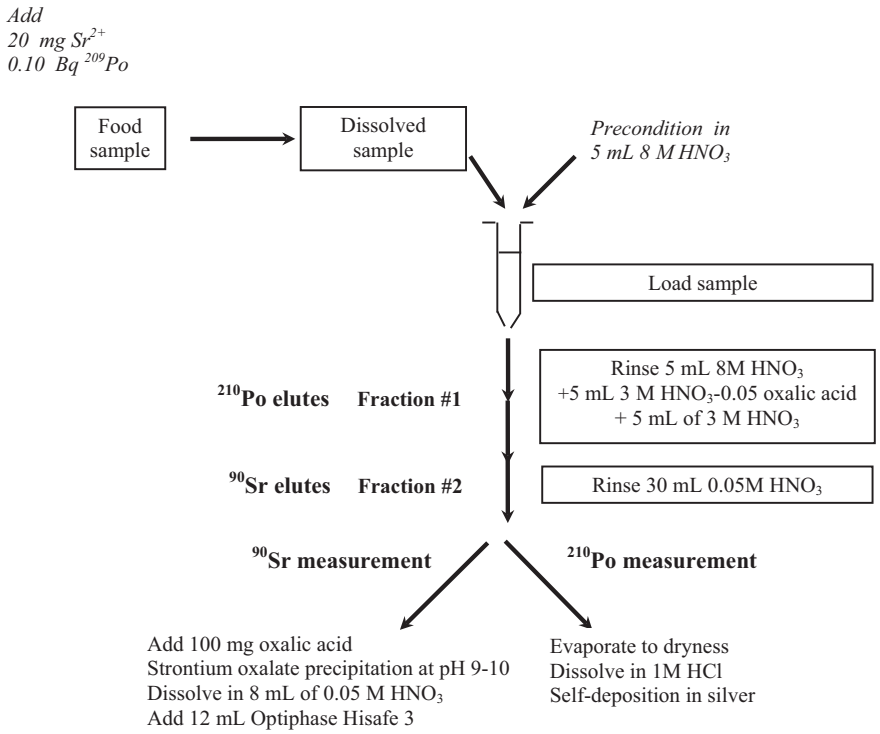

Fig. 1. Radiochemical procedure for ^{90}Sr and ^{210}Po isolation using prepacked Sr resin columns.

microwave digestion that only is able to accept 0.5–1 g of organic sample, and gets a better quantification limit L_Q (see Section 3.2.). In order to evaluate ^{90}Sr and ^{210}Po recovery, stable strontium carrier (20 mg Sr^2) and ^{209}Po internal standard (0.1 Bq) were added. Then the sample aliquot was leached following EPA 3050B method (USEPA., 1996) with several additions of nitric acid, HNO_3 (first, 1:1 and next concentrated) and hydrogen peroxide, H_2O_2. The first addition of nitric acid was gradually heated otherwise it reacts vigorously producing foam. Next the solution was passed through a mixed cellulose membrane filter with 0.45 μm pore diameter (Millipore), and heated to dryness. Afterwards, it was re-dissolved with 10 mL of 8 M HNO_3. A simple inspection shows that this step provides full sample digestion of the sample aliquot.

Step 2. *Isolation of strontium and polonium using Sr resin.* Previously 5 mL 8 M HNO_3 were passed through the resin column in order to precondition it. Then the sample solution obtained in Step 1 was immediately loaded and eluted from the resin column (Fraction #1). In order to isolate polonium, 5 mL of 8 M HNO_3 followed by 5 mL of 3 M HNO_3 – 0.05 oxalic acid, and 5 mL of 3 M HNO_3, were successively loaded to the resin column. Furthermore strontium is highly retained in nitric acid concentration by the resin column (Philip Horwitz, Chiarizia, & Dietz, 1992). Finally, strontium was separated loading 3 × 10 mL 0.05 M HNO_3 (Fraction #2). The column may be regenerated adding 10 mL of deionised water and stored in 0.05 M HNO_3.

Step 3. *Preparation of the sources for measurement.* The fraction containing polonium (Fraction #1) was evaporated to complete dryness and the obtained residue dissolved in 1 mL of concentrated HCl acid in order to change to hydrochloric media. This evaporation step was repeated three more times and the final residue was dissolved in 50 mL 1 M HCl. Silver discs were utilized to polonium self-deposition during 6 h in 1 M HCl at about 80 °C (Vajda, LaRosa, Zeisler, Danesi, & Kis-Benedek, 1997). The addition of few milligrams of ascorbic acid avoids iron plating on the silver foil. Additionally the self-deposition process also isolates polonium from other potential interferences and makes it ready to be measured via alpha-particle spectrometry. The polonium peaks have high resolution in the alpha spectra (less than 35 keV in all cases), revealing an outstanding performance of the self-deposition step, without thick deposits.

Regarding strontium determination 100 mg of oxalic acid were added to Fraction #2. The precipitation of strontium oxalate in Fraction #2 was made possible by raising pH to 9–10 with NH_3OH. The strontium precipitate was filtered and its chemical recovery was determined through gravimetric measurement (Mosqueda, Villa, Vaca, & Bolívar, 2007). Then the strontium precipitate was dissolved in 8 mL of 0.05 M HNO_3 and placed into a polyethylene vial. Next 12 mL of Optiphase Hisafe 3 were added to the solution and immediately it was vigorously shaken during a few minutes. Finally, vials were kept into the liquid scintillation counter during 2 weeks until reach ^{90}Sr/^{90}Y secular equilibrium (approximately 2–3 weeks).

3. Results and discussion

3.1. Optimization of the method

3.1.1. Alpha-particle spectrometry

The proposed method is based on isotope dilution alpha spectrometry. ^{209}Po tracer was added to the food samples in order to quantify the radiochemical yield and the ^{210}Po activity. The decay corrections were appropriately applied to the calculation of ^{210}Po and ^{209}Po activities. High chemical yields (>50%) were achieved in the determinations carried out by alpha-particle spectrometry (see Table 1).

Finally, several blank samples were measured during a background counting time of 3 days in order to check the quantification limit (see Section 3.2.).

3.1.2. Liquid scintillation counting

The LSC measurements were carried out using polyethylene vials instead of glass vials because of their lower scintillation background (Rapkin & Gibbs, 1963).

Table 1
^{90}Sr and ^{210}Po activity concentrations in seafood samples collected at the Mediterranean coast of Spain. 1 g samples were processed. The uncertainties σ are expressed as k = 1.

Food type	A(^{90}Sr) (Bq kg^{-1})	σ (k = 1)	Chemical recovery %	A(^{210}Po) (Bq kg^{-1})	σ (k = 1)	Chemical recovery %
Sea urchin	<L_Q		56%	56	11	75%
Oyster	<L_Q		60%	934	41	59%
Mussel	<L_Q		59%	172	9	80%
	<L_Q		69%	354	17	74%
	<L_Q		53%	256	12	85%
	<L_Q		64%	390	14	76%
	<L_Q		67%	397	15	88%
	<L_Q		70%	224	10	64%
	<L_Q		75%	91	3	68%

The influence of sample solution/scintillation cocktail ratio on the background and the counting efficiency was studied. The total volume in the vial was kept at 20 mL.

First, the background count rate was measured varying the volume of 0.05 M HNO_3 (from 2 to 8 mL) added to the scintillation cocktail (see Step 3 in Section 2.5.). The measurement time was set to 600 min. and the obtained results are shown in Table 2.

Second, the counting efficiency for ^{90}Sr–^{90}Y was obtained through tracing twenty solutions with known quantities of ^{90}Sr–^{90}Y. Twenty polyethylene vials were traced with about 0.5 g of ^{90}Sr–^{90}Y standard solution and 100 mg oxalic acid dissolved in different volumes of 0.05 M HNO_3. The counting time was set to 600 min. The counting efficiency for ^{90}Sr–^{90}Y was calculated using the following equation:

$$\varepsilon = \frac{n_T - b}{M \cdot A} \quad (1)$$

where n_T is the total count rate; b the background count rate; A the mass-related activity concentration of the ^{90}Sr–^{90}Y standard solution (Bq g^{-1}), and M its mass (g) added. The obtained results are shown in Table 2.

Finally, the background count rate and the counting efficiency are combined into the figure of merit (FOM) defined as follows (Knoll, 2010):

$$FOM = \frac{\varepsilon^2}{b} \quad (2)$$

where b is the background count rate (cpm), and ε is the counting efficiency.

In view of the results (see Table 2), the volume of the sample solution was fixed to 8 mL because it provides the maximum value of FOM.

Quench was quantified with the standard quenching parameter SQP(E) (Villa et al., 2003; Villa et al., 2007). The SQP(E) parameter is based on the interaction between the radiation from an external ^{152}Eu radioactive source and the scintillator mixed with the sample in the polyethylene vial. When quenching effect is increased SQP (E) is reduced. The average SQP(E) value was 690 ± 3 for samples and 692 ± 3 for an unquenched blank. We conclude, therefore, that no differences in quench were found in samples following our procedure (Kim, Kim, & Lee, 2001).

In a previous paper the PSA threshold of the LSC system was optimized and set to 105 (Villa et al., 2003). In this way the alpha interferences into the ^{90}Sr beta window were reduced.

3.2. Validation of the method

Firstly, the method was validated in terms of the quantification limit (L_Q), defined as (Currie, 1968):

$$L_Q = \frac{50\{1 + \sqrt{1 + b/12.5}\}}{\varepsilon \cdot \tau \cdot M \cdot R} \quad (3)$$

where b is the total background counts; τ the background counting time (in seconds) which, in this case, is the same as the sample counting time; ε is the counting efficiency; R the radiochemical yield; and M is the mass of the sample.

The results are shown in Table 2 (in Bq kg^{-1}, assuming 1 g of sample). The calculated L_Q values were lower than the maximum permissible levels as presented in EU Council Regulation 2016/52/Euratom.

Then the IUPAC (Thompson, Ellison, & Wood, 2006) and ISO (ISO/IEC., 2010) recommendations for assessment of performance of laboratories were applied to the results obtained. More specifically a z-score test, u-test, and trueness and precision tests were performed for the determination of ^{90}Sr and ^{210}Po in three types of IAEA certified reference materials, with activities ranging over 2 orders of magnitude. Five aliquots of 1 g were analyzed for each CRM by using the proposed method (N = 5) and results are shown in (Table 3). The z-score and u values obtained are inside the acceptable range for trueness of the proposed method. All the results obtained an "Acceptable" status referring to precision and trueness. The analysis of the IAEA-437 sample gives a ^{210}Po activity of 4.5 Bq kg^{-1}, very close to the reference material reported value and within its 95% confidence interval. Referring to IAEA-330 sample, the median value obtained with the proposed method is 18.4 Bq kg^{-1}, which is within its 95% confidence interval. Lastly for IAEA-414 sample the obtained median value is within the reported 95% confidence interval for both radionuclides. However, the precision of the laboratory performance for ^{90}Sr determination is very high (Table 3) due to the fact that the uncertainty associated to the information value is also very high.

Table 2
Quenching parameter SQP(E), counting efficiency (ε), background count rate (b), Figure of Merit (FOM) and quantification limit (L_Q), obtained for ^{90}Sr and different volumes of 0.05 M HNO_3, being 600 min the counting time in Quantulus detector and assuming 1 g of sample analyzed. The uncertainties σ are expressed as k = 1.

Volume 0.05 M HNO_3 (mL)	SQP(E)	ε	σ (k = 1)	b (cpm)	σ (k = 1)	FOM ε ^{2}/b (cpm^{-1})	L_Q (Bq kg^{-1})
2	748	0.922	0.009	7.9	0.4	0.11	26.4
4	738	0.918	0.010	6.3	0.3	0.13	26.2
6	706	0.926	0.008	6.4	0.3	0.13	25.8
8	690	0.893	0.009	5.3	0.3	0.15	25.3

Table 3
Results obtained for three IAEA reference materials and through a national intercomparison exercise (CSN) for five aliquots (N = 5). Uncertainties (σ) are expressed at k = 1.

ID	Radionuclide	A_{LAB}		A_{REF}		z-score	u	Trueness	Precision P (%)
		Bq kg^{-1}	σ (k = 2)	Bq kg^{-1}	σ (k = 2)				
IAEA-437 (mussel)	^{210}Po	4.5	0.3	4.2	0.7	0.4	0.4	Acceptable	18
	^{90}Sr	–	–	–	–	–	–	–	–
IAEA-330 (spinach)	^{210}Po	–	–	–	–	–	–	–	–
	^{90}Sr	18.4	0.2	20.10	2.00	-0.8	0.8	Acceptable	11
IAEA-414	^{210}Po	2.4	0.3	2.1	0.3	1	0.7	Acceptable	19
(fish)	^{90}Sr	0.35	0.05	0.28	0.2	0.4	0.3	Acceptable	72
CSN	^{210}Po	109	5	102	2	3.3	1.3	Acceptable	5
(organic ashes)	^{90}Sr	74.5	1.3	70	2	2.3	1.9	Acceptable	3

The observed deviations are in the order of the precision values associated with environmental materials, and overall, we conclude that the proposed method is adequate to analyse ^{210}Po and ^{90}Sr in food samples.

Finally, our laboratory took part in an intercomparison programme with environmental radioactivity laboratories (2010) organized by the CSN (Nuclear Security Spanish Council) to measure ^{90}Sr and ^{210}Po in ashed food samples. In this exercise the activity concentration obtained by our laboratory was 109 ± 5 Bq kg^{-1} and 74.5 ± 1.3 Bq kg^{-1} respectively for ^{210}Po and ^{90}Sr (Table 3). The reference values given were 102 ± 2 Bq kg^{-1} and 70 ± 2 Bq kg^{-1} respectively for ^{210}Po and ^{90}Sr. The obtained results are in agreement with the certified ones within the uncertainty calculated.

The uncertainty analysis of the obtained results was done using the GUM Workbench software (GmbH, 2016) recommended from BIPM (JCGM 100., 2008), concluding that uncertainty from counting statistics and the mass of the stable Sr tracer were the largest contributors to measurement uncertainty, accounting for approximately 84% and 16%.

3.3. Determination of ^{210}Po and ^{90}Sr in real samples

After the validation procedure, the radiochemical method described in this work was applied to different types of seafood and its routine applicability for controls is evaluated (Section 2.4.). The following types of samples were analyzed: sea urchin, bivalve molluscs (M. galloprovincialis), and oyster. The results obtained are reported in Table 1.

The dry weight concentration of ^{210}Po in the samples varied between 56 and 934 Bq kg^{-1}. The lowest activity concentration was found in the sea urchin, this value is sixteen times lower than the one measured in the oyster sample, another common seafood in Spain that reached the highest value.

The ^{90}Sr activity concentration in all of the analyzed samples was below the quantification limit (<30 Bq kg^{-1}). No ^{90}Sr was detected in any of the seafood samples analyzed.

4. Conclusion

^{210}Po and ^{90}Sr were sequentially extracted from food samples by developing a precise, accurate and reliable procedure through liquid scintillation counting and alpha-particle spectrometry. A selective detection of ^{210}Po and ^{90}Sr and also a faster radiochemical procedure was provided by the developed chemical method. Moreover, a low quantification limit was obtained, 25 Bq kg^{-1} and 2 Bq kg^{-1} for ^{90}Sr and ^{210}Po respectively. Finally, the proposed method is especially suitable for food safety monitoring due to an effective pre-treatment of samples.

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