Applied Radiation and Isotopes 93 (2014) 29–32

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Quality control assurance of strontium-90 in foodstuffs by LSC

I. Lopes *, A. Mourato, J. Abrantes, G. Carvalhal, M.J. Madruga, M. Reis

Instituto Superior Técnico, Campus Tecnológico e Nuclear, Laboratório de Proteção e Segurança Radiológica, Estrada Nacional 10 (ao km 139,7), Bobadela LRS 2695-066, Portugal

HIGHLIGHTS

- Blank and background samples were analyzed according to internal quality control program established.
- Analysis of certified reference material was carried out to perform additional evaluation of the accuracy.
- Analysis of interlaboratory comparison sample showed *Z*-score and *Z* (*ML*) acceptable values.
- The Sr-90 activity measured in wild bilberry powder sample was in agreement with the reference value.

ARTICLE INFO

Available online 7 February 2014

Keywords:
Quality control
Strontium-90
Foodstuffs
Liquid scintillation

ABSTRACT

A method based on the separation of Sr-90 by extraction chromatography and beta determination by Liquid Scintillation Counting (LSC) technique was used for strontium analysis in food samples. The methodology consisted in prior sample treatment (drying and incineration) followed by radiochemical separation of Sr-90 by extraction chromatography, using the Sr-resin. The chemical yield was determined by gravimetric method, adding stable strontium to the matrix. Beta activity (Sr-90/Y-90) was determined using a low background liquid scintillation spectrometer (Tri-Carb 3170 TR/SL, Packard). The accuracy and the precision of the method, was performed previously through recovery trials with Sr-90 spiked samples, using the same type of matrices (milk, complete meals, meat and vegetables). A reference material (IAEA_321) was now used to measure the accuracy of the procedure. Participation in interlaboratory comparison exercises was also performed in order to establish an external control on the measurements and to ensure the adequacy of the method.

1. Introduction

Because of its long physical and biological half-lives, Sr-90 is one of the most hazardous radionuclides and it may cause damage to the bone marrow (UNSCEAR, 2000). It undergoes β^- decay into Y-90 and both were released together to the environment, due to nuclear weapons testing in the atmosphere and from the nuclear fuel cycle. Immediately following a nuclear accident, the fresh fallout contains others radionuclides together with high radiostrontium activity ratio (Sr-89/Sr-90). The old fallout (several years after release) contains only beta emitter Sr-90 ($T1/2=28.5$ years) and its daughter Y-90 ($T1/2=64.4$ h). For these reason, the strontium levels in the environmental samples, with particular emphasis on foodstuffs, are of particular concern (Groska et al., 2012; Spasova et al., 2008).

Radiochemistry methods for the determination of Sr-90 in environmental samples require complex techniques for strontium to be separated from the sample matrix and from other interfering radionuclides (Vajda and Kim, 2010; Kim et al., 2009). The Sr-90 and its daughter Y-90 must also be separated from the sample matrix prior to measurement. In case of environmental samples containing low levels of Sr-90, large amounts of sample are often required which makes sample processing not only more labor-intensive but also required more sensitive methods. By solid phase extraction, using a strontium specific resin (Sr-resin, Eichrom), Sr-90 can be isolated without interferences from other radionuclides (Lee et al., 2013). The Sr-resin is specific to Sr ions and enables rapid and simple column chromatographic separation of strontium from calcium, potassium and many other elements with of 3–8 mol l^{-1} HNO_3 solution (Grahek et al., 2011). The determination of Sr-90 can be performed by measuring the radioactivity of either Sr-90, Y-90 or of both if the degree of secular equilibrium is known (Karacan, 2011). The beta measurements can be performed using liquid scintillation spectrometers or proportional counters (Popov et al., 2009). Although the background of liquid scintillation spectrometers are higher than that of proportional counters, liquid scintillation is favored due to the much better spectral resolution.

In general, appropriate procedures and validated methods should be used, following the requirements specified in ISO/IEC

* Corresponding author.
E-mail address: ilopes@ctn.ist.utl.pt (I. Lopes).

http://dx.doi.org/10.1016/j.apradiso.2014.01.022
0969-8043

17025 international standard (ISO/IEC, 2005). The validation is the process used to confirm that the analytical procedure employed for a specific test is suitable for its intended use and should be as extensive as necessary to meet the needs of the given application. The methods need to be validated or revalidated, whenever the conditions for which the method has been validated changed (e.g., scope of the method, different matrix, instrument with different characteristics) (Drábová et al., 2012). The QC programs (internal and external) should include, among others, all necessary actions to assure if the equipment is calibrated and operating satisfactorily, to verify specified requirements such as accuracy and precision, and to detect potential faults in routine measurements, and should also include regular participation in interlaboratory comparison exercises.

Results of validation trials using spiked samples were already reported (Lopes et al., 2010). The aim of this paper is to evaluate the accuracy of the method using a reference material and interlaboratory comparison samples.

2. Experimental

The analytical method applied for Sr-90 determination in foodstuffs has been described elsewhere (Lopes et al., 2010). The method is based on the digestion of the sample and separation of strontium by extraction chromatography, using the Sr-resin. About 7 g of ashes were taken for analysis. During the digestion of the sample, stable strontium was added (≈ 20 mg) to determine the chemical yield gravimetrically, and the mixture was heated to boiling and filtered. The Sr-90 was separated by oxalate and carbonate precipitations. Afterwards, the residue was dissolved with 3 mol l^{-1} HNO_3 and loaded onto a 20 ml column filled with Sr- resin (100–150 μm), pre-rinsed with 3 mol l^{-1} HNO_3. The column was rinsed first with 8 mol l^{-1} HNO_3 followed by 3 mol l^{-1} HNO_3 and the strontium retained in the column was stripped with 0.05 mol l^{-1} HNO_3. After evaporation to dryness, the residue was dissolved with 8 ml of 0.1 mol l^{-1} HCl and mixed with 12 ml of Ultima Gold LLT cocktail (Perkin Elmer) in a glass scintillation vial. The measurements were performed by Liquid Scintillation Counting (LSC) technique using Packard Model Tri-Carb 3170 TR/SL spectrometer. The detection system was calibrated by measuring Sr-90/Y-90 standard sources, and using CCl_4 as quenching agent. The procedure was also adapted for small columns (2 ml) when the amount of ash sample was not enough to perform the analysis.

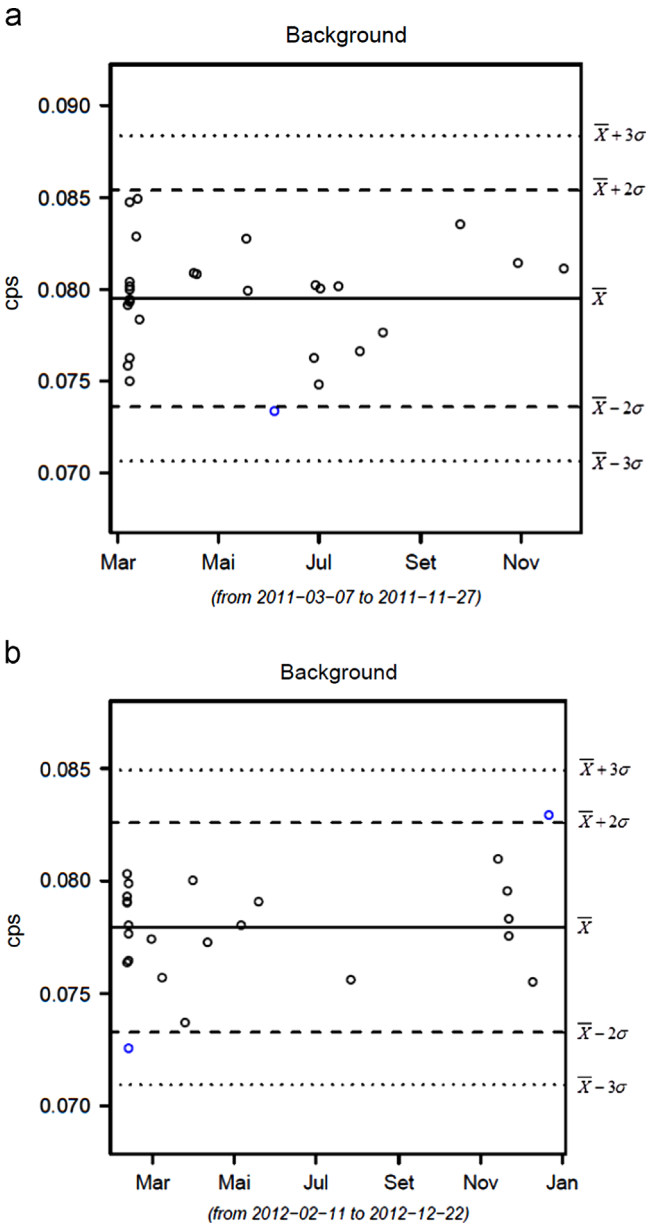

Fig. 1. Background control charts (Years: 2011 (a); 2012 (b)).

3. Results

3.1. Internal quality control

Blank and background samples were analyzed, according to internal quality control program. A background sample was always measured with each set of 10 samples (Fig. 1) within each batch and analyses of blank samples were also performed (Fig. 2) for each batch. Control charts were used to help in the identification of possible systematic deviation from regular performance in time, for inspection of trends and to verify the stability of the counter. The average background counts ($\overline{X}$) was calculated, the standard deviation was used to set the warning ($\overline{X} \pm 2\sigma$) and actions limits ($\overline{X} \pm 3\sigma$), respectively, considering the first background measurements carried out during the calibration. For blank samples, the control chart was established at calibration date but the limit lines are recalculated for all plotted data. The equipment was operated under controlled room temperature conditions. Calibration of the LSC spectrometer is carried out once per year using Sr-90/Y-90 standards prepared from a certified standard solution (Amersham) with increasing amounts of CCl_4 (quenching agent) allowing the determination of the calibration curve (Quench Indicating Parameter not constant).

The accuracy and the precision were determined during the validation trials, using Sr-90/Y-90 spiked samples, prepared from a mixture of several foodstuffs samples ashes (Lopes et al., 2010). The precision was estimated to be less than 4% and a good agreement between the added and the measured activities was obtained (the recovery percentages ranged from 70 to 80%).

In routine work, replicates samples are not performed regularly since the amount of ashes obtained from the samples incineration is not enough to perform duplicate and/or triplicate samples. Furthermore, since the Sr-90 activity levels were below the detection limits, is not possible, in each batch, evaluate the precision of the procedure.

Analysis of a certified reference material was also carried out, in order to perform additional evaluation of the accuracy for the validation of the method.

The reference material (IAEA_321) used to verify the accuracy of the method was a milk powder collected from European