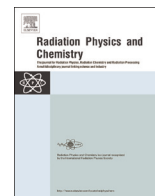




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Environmental radioactivity measurements and applications – Difficulties, current status and future trends

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HIGHLIGHTS

- Determination of γ -emitting radionuclides in environmental samples is reported.
- The use of radionuclides as tracers of environmental processes is reviewed.
- Detection limits improvement will allow for new uses of radionuclides as tracers.
- Radionuclide size fractionation in environmental materials should be investigated.
- Radioactive equilibrium in environmental materials should be further investigated.

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ABSTRACT

For several decades natural and artificial radioactivity in the environment have been extensively studied all around the world. Nuclear accidents – mainly that of Chernobyl – have led to the development of the field of radioecology, while detector systems and techniques – with predominant that of γ -spectrometry – have been continuously developed through the years to meet researchers' needs. The study of natural radionuclides that was originally limited to ^{226}Ra , ^{232}Th and ^{40}K was then extended to include radionuclides such as ^{234}Th , ^{210}Pb , ^{235}U and ^7Be , which allowed the study of radioactive equilibrium.

Besides their importance from the radiation protection point of view, many radionuclides are also used as tracers of environmental processes, such as aerosol and transportation of air masses studies (^7Be , ^{10}Be , ^{22}Na), soil erosion, sedimentation and geochronology (^{210}Pb , ^{137}Cs), marine ecosystems studies and studies related to climate change. All these studies require specialized samplings strategies and sampling preparation techniques as well as high quality measurements, while the improvement of detection limits is often of vital importance.

This work is a review of environmental radioactivity measurements and applications, mainly focused in the field of γ -spectrometry, for which difficulties and limitations will be presented, together with future trends, new challenges and applications.

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1. Introduction

Radioactivity in the environment has been extensively studied around the world for more than 50 years. Nuclear accidents with predominant that at Chernobyl, due to their global scale have motivated scientists to extend their research, improve analytical techniques and develop new, to detect and quantify radionuclides posing a threat to human, thus resulting to the development of the field of radioecology.

Among the techniques most often used for the analysis of environmental radioactivity are γ -spectrometry, α -spectrometry and liquid scintillation. Since α -spectrometry requires radiochemical

preparation and is time and labor consuming, it has not been so widely used for environmental radioactivity studies, compared to γ -spectrometry, which has been the workhorse of environmental radioactivity analysis. Liquid scintillation on the other hand has clearly an advantage for the analysis of liquid samples.

The radionuclides that were first given much attention in environmental materials – mainly soil, fossil fuels and ashes from their combustion – are ^{226}Ra , ^{232}Th and ^{40}K (Anagnostakis et al., 1996), since they were considered as the major contributors to the human dose, and the first formula for the estimation of the external γ -ray dose rate due to terrestrial radiation was based on these three radionuclides (UNSCEAR, 1993). It should be mentioned that, though ^{226}Ra is not a significant γ -emitter itself and ^{232}Th does not emit photons at all, they were considered as good

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representatives of the radioactive series of ^{238}U and ^{232}Th respectively. Another reason for the interest on ^{226}Ra was radon (^{222}Rn) exhalation. Other radionuclides such as ^3H , ^7Be and ^{14}C and ^{210}Pb were also reported, though not so often, one reason being the difficulties in their detection.

As the detector technology advanced and new detector types such as LEGe detectors were made available, the determination of radionuclides emitting low energy photons, such as the decay products of ^{238}U ; ^{234}Th (63.29 keV) and ^{210}Pb (46.52 keV) was made possible. This showed that radioactive equilibrium – especially between ^{226}Ra and ^{210}Pb – might be significantly disrupted in environmental materials such as soil (Anagnostakis et al., 2005; Dowdall and O’Dea, 2002) and NORM (Karangelos et al., 2004a). Another example of interesting behavior in the environment and equilibrium disruption is between ^{210}Pb and its short-lived α -decay product ^{210}Po (Persson and Holm, 2011).

The radionuclides of the ^{235}U series in environmental materials were not paid much attention in the past, due to (i) the low activity and importance from the radiation protection point of view, compared to ^{238}U and ^{232}Th series radionuclides, (ii) the assumption of natural isotopic abundance of uranium and (iii) the difficulty in the ^{235}U determination with γ -spectroscopic techniques. However, the use of depleted uranium (DU) in military conflicts and the subsequent DU soil contamination, demonstrated the need for independent ^{235}U determination in environmental materials and today there are such reports available (Uyttenhove et al., 2002; Karangelos et al., 2004b).

The last few years there is a lot of interest in the determination of cosmogenic radionuclides – primarily ^7Be – in various types of environmental samples, mainly atmospheric aerosol and water. Other radionuclides studied today in atmospheric aerosols are ^{22}Na , ^{210}Pb , ^{40}K and ^{137}Cs . Furthermore many reports exist on the use of radionuclides in the study of environmental processes in the ground, water and in the atmosphere, while medical applications as sources of radioactivity in the environment are currently under study too (Fischer et al., 2009).

2. Gamma spectrometry in the analysis of environmental samples

Though γ -spectrometry is a well established and widely applicable analytical technique for the determination of γ -emitting radionuclides, the determination of natural and artificial radionuclides in environmental samples may require some special considerations to ensure quality results. Detector calibration, use of correction factors, detector background, detection limits and interference between photopeaks are some of them.

2.1. Detector calibration

A common way of detector calibration in gamma spectrometry is experimentally; using certified mixed radionuclide solutions containing a series of radionuclides, to cover the energy region 59–1836 keV, where most of the radionuclides usually found in the environment emit photons.

The last few years an extensive use of computational methods – often combined with experimental methods – for efficiency calibration is observed. Progress in computational calibrations was mainly the result of development of powerful computers – including parallel processing – and user-friendly graphical interfaces. Two types of computer codes are used for computation calibrations:

- (i) General-purpose Monte Carlo (M-C) simulation computer codes, like MCNP, PENELOPE, GEANT, ETRAN etc., that can be

used for detector calibrations. Among the results given by these codes is the probability of a photon to deliver in the detector energy within a specific energy window (bin). By selecting the appropriate bin width, taking into consideration the detector energy resolution, the user obtains results useful for the determination of full energy peak efficiency. Incorrect bin width selection may introduce significant type-B uncertainty, especially for lower energy photons in bulk sources (Karfopoulos and Anagnostakis, 2010). Another issue that has to be taken into consideration during simulation is the detector geometric characteristics. Though, most of them are known to the user, the dead layer thickness needs to be determined. For this purpose an iterative method that is based on the comparison of experimental measurements of efficiency and simulation results (Agrafiotis et al., 2011) has to be followed. The type-B uncertainty introduced by this procedure should not be neglected.

- (ii) Dedicated computer codes for detector calibrations. Several such codes have been developed the last few years for the calibration of Ge-detectors; some of them are based on analytical calculations, others on M-C simulation and others on mixed solutions. Besides the efficiency calibration some of the codes are also calculating correction factors for self-absorption or true coincidence. A list of such codes includes ANGLE, CCCC, CORSUM, GESPCOR, ETNA, EFFTRAN, ISOCS, etc.

2.2. Correction factors

The most important corrections that have to be made in γ -spectroscopy are self-attenuation corrections, true coincidence and random summing corrections.

Self-attenuation is the attenuation of photons within the sample, and it is very important for the low activity environmental materials due to the use of large volume samples. It depends on the sample thickness, photon energy and material type and density, and is therefore different between calibration standard and sample. To take into consideration for this difference an efficiency correction factor η must be used (Anagnostakis and Simopoulos, 1996). Efficiency corrections for such materials are usually required for photons below 200 keV, such as those emitted by ^{234}Th (63.29 keV) and ^{210}Pb (46.52 keV). For the 46.5 keV photons emitted in a 7 cm thick cylindrical vessel containing soil, η may be less than 0.5. For very dense materials or NORM samples from oil exploitation corrections may be needed for much higher energies (Landsberger et al., 2013).

Various techniques have been developed for efficiency correction calculations. Some of them require experimental work, such as that proposed by Cutshall et al. (1983) which is based on the measurement of the attenuation in the sample of photons from an external source, to obtain the self-attenuation of the photons inside the sample. This correction gives acceptable results for a sample thickness ~ 30 mm. For soil samples of 4 mm thickness this method overestimates η by about 9% (Jodłowski et al., 2014). Another technique for self-attenuation correction is based on the calculation of an integral giving the efficiency of the detector for both the calibration standard and the sample (Debertin and Helmer, 1988). For this calculation the linear attenuation coefficients μ for the calibration standard and the sample are required. This is a drawback of the technique, due to lack of data for μ of materials of environmental interest. Experimental data of μ for environmental materials may be found in Petropoulos et al. (2002), while computer codes for μ calculation for materials of known composition have also been developed (Jaegers and Landsberger, 1990).

The second correction that has to be applied is for true coincidence – the co-detection of two photons emitted from the decay

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