



Possibilities of the use of CeBr₃ scintillation detectors for the measurement of the content of radionuclides in samples for environmental monitoring

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ABSTRACT

The investigation of radioactivity in samples is an application of gamma-ray spectrometry dealing with low and very low level gamma-ray activities of different isotopes. Gamma-ray spectrometry performed in the framework of radiological environmental monitoring may be done after selective sampling processes or after a chemical purification of a sample. Both cases imply that only some specific radionuclides should contribute to the obtained spectrum. Gamma-ray spectrometry performed with medium energy resolution detectors may allow the possible distinction of their photopeaks. Therefore, a cerium bromide (CeBr₃) detector can be particularly attractive for routine tasks in radiological environmental monitoring as it has a high efficiency, medium energy resolution and it can work at room temperature. This study describes the conditions under which a CeBr₃ detector can serve for some routine analysis in radiological analysis of samples collected in the environment or collected by air-samplers in environmental radiological monitoring programmes.

1. Introduction

Because of their high energy resolution, hyperpure germanium detectors (HPGe) are routinely chosen with the aim of obtaining high quality gamma-ray spectra from any type of matrix for any gamma-ray analysis objective. However, these detectors need to be cooled to very low temperatures. Therefore, they need a continuous supply of liquid nitrogen or an electric cooling, adding upfront and operational costs and complexity to the system. That implies that the use of these detectors becomes expensive. Additionally, with both methods, time for cooling is required before use.

In recent years there has been a growing interest in the development of new scintillator materials, promoted by the increasing number of industrial, medical and scientific applications looking for scintillation crystals with high luminosity, short decay time, high density and low costs for purchase and maintenance for the use in gamma-ray analysis.

Bromide scintillators, like CeBr₃, can be particularly attractive as they meet those requirements. In particular, CeBr₃ is a crystal of interest in the pursuit of medium to high energy resolution gamma-ray spectrometry at room temperature (Shah et al., 2004; Guss et al., 2009). Its resolution is better than that of a typical NaI(Tl) detector, but worse than a HPGe detector.

Of course, the adequacy of a detector for a specific purpose depends

not only on the detector characteristics, but also on the client requirements, which include the radionuclides to deal with and the uncertainties and detection limits desired. These boundary conditions depend on the ability of the detector to separate close photopeaks, i.e. its resolution, its efficiency, its internal background and the software used to analyze the spectrum. Concerning laboratory measurements, it also depends on the measurement characteristics: shielding, sample geometry and counting time.

Therefore, in the last few years some publications have investigated CeBr₃ detector characteristics, some of them focused on the crystal properties (Quarati et al., 2013) or on applications as a high resolution gamma-ray detector (Guss et al., 2009) and others on its application to specific uses such as early warning networks (Glavic-Cindro et al., 2017) or radionuclide assessment in NORM materials (Peyres et al., 2017) or in sediment samples (Androulakaki et al., 2020). From these papers it is observed that a CeBr₃ detector can also achieve a relatively high full-energy peak efficiency for gamma spectrometry, which enables it to be used in low-level activity applications such as the monitoring of radioactivity in the environment done in laboratories with collected samples.

Gamma-ray spectrometry dealing with low and very low level gamma-ray activities of different samples is a fundamental part of radiological environmental monitoring programmes. The medium

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resolution of CeBr₃ detectors could be sufficient to satisfy the quality requirements (uncertainties and detection limits) concerning the determination of the main radionuclides usually required in those programmes for in-lab measurements of collected samples, like a quantitative activity concentration measurement of ¹³¹I and ¹³⁷Cs and radium isotopes by gamma-ray spectrometry. These detectors will allow for an easier and cheaper usage than the conventional HPGe detectors.

As well as these, one needs to remember that part of the laboratory gamma-ray spectrometry done in the frame of radiological environmental monitoring on collected samples is done after selective sampling processes – like the one done by using filter cartridges for the collection of iodine – or after nuclide chemical isolation – like some radium determination procedures. Both cases imply that only some specific radionuclides should appear in the obtained gamma-ray spectrum from the investigation of such samples, so that this medium energy resolution is enough to distinguish their gamma-ray lines. Also, the activity concentration of samples that contain more radionuclides, typically those belonging to the natural radioactive decay chains, can be measured by using these CeBr₃ detectors if the radionuclides we want to analyze, having activities above detection limits, do not present strong overlapping with those belonging to other radionuclides present in the sample or those present in the background signal.

In this paper, and in order to address the adequacy of this type of detectors for laboratory measurements in radiological environmental monitoring, we have tested the suitability of a cylindrical 1.5" x 1.5" CeBr₃ low background detector (crystal diameter = 1.5" and crystal length = 1.5"), shielded by 10 cm thick lead, for the investigation of different test samples (different matrices in specific geometries) from collected samples in monitoring programmes and for radionuclides usually assessed in these programmes: ¹³¹I, ¹³⁷Cs and Ra isotopes. After carrying out appropriate calibrations in efficiency and resolution for those samples, the activity concentration of real samples has been measured and, using the Gamma Vision software to process the spectra obtained, uncertainties and detection limits for some radionuclides have been obtained. Afterwards, the ability of these detectors to achieve the regulatory detection limits, in our case defined by the Spanish Nuclear Safety Council (CSN, 1993) and by EURATOM (Council Directive, 2013), is checked, highlighting the conditions in which they can be incorporated into environmental radioactivity monitoring programs, in our case in the National Sampling Stations Network, instead of the HPGe detectors.

Finally, the validity of the results obtained has been checked by comparison to reference values employing different reference materials.

2. Materials

2.1. Equipment

A 1.5" x 1.5" CeBr₃ detector (crystal volume = 43.4 cm³) from Scionix (low contamination crystal, with less than 0.008 intrinsic counts/s/cm³), with a 2" Hamamatsu photomultiplier, has been used. It has a nominal energy resolution of 4.3% at 662 keV (¹³⁷Cs). It has a 0.5 mm thick Al metal body housing and a 45 mm outside diameter. The whole system (detector and photomultiplier) and sample are contained in a 10 cm thick Pb shield.

For the digital multichannel analyzer, 512 channels have been selected according to the suggestion by Knoll (2010) of allocating around 4–5 channels across the FWHM (Full Width at Half Maximum), on the basis of a constant channel width. For an energy interval between 60 and 1836 keV, this selection implies an acceptable value of FWHM at 200 keV (≥4–5 channels per FWHM). 200 keV is the threshold of the energy range of interest (¹³¹I, ¹³⁷Cs and Ra gamma emitting isotopes).

Detector calibration was made by means of different calibration sources prepared by using a mixed nuclide certified reference solution from Eckert&Ziegler (with radionuclides shown in Table 1).

Table 1

List of radionuclides used for the calibration of the CeBr₃ detector together with their energies and the FWHM observed for them. The FWHM observed with a 40% coaxial HPGe detector is also shown.

Radionuclide	Energy in keV	FWHM in keV	
		1.5" x 1.5" CeBr ₃	40% HPGe
²⁴¹ Am	59.54	8.02	0.76
¹⁰⁹ Cd	88.03	10.27	0.79
⁵⁷ Co	122.06	11.59	0.84
¹³⁹ Ce	165.86	13.61	0.88
²⁰³ Hg	279.2	18.94	1.02
¹¹³ Sn	391.7	22.64	1.13
⁸⁵ Sr	514	25.85	1.20
¹³⁷ Cs	661.66	30.75	1.33
⁸⁸ Y	898.04	35.11	1.54
⁶⁰ Co	1173.23	38.76	1.75
⁶⁰ Co	1332.49	42.66	1.86
⁸⁸ Y	1836.05	48.56	2.22

2.2. Samples

The detector was tested by using different types of test samples, prepared using different environmental matrices contained in typical containers commonly used in radiological environmental monitoring. Eight different types of test samples from air, water, milk and soil matrices were tested. In three of these eight cases, ¹³¹I and radium isotopes are extracted from the original matrix.

To obtain the air test samples, two sampling systems were used. The first one, a system that circulates 300 m³ of air per week, through a radioiodine collection filter cartridge (activated charcoal filter, RADeCO model CP-100 47 mm effective diameter and 26 mm height) and a plain cellulose nitrate membrane filter (particulate filter, Whatman 7188-004 WCN, pore size: 0.8 µm, diameter: 47 mm but 40 mm effective diameter and thickness: 0.1 mm). In this case, both the cartridge and the filter were test sampled weekly, as well as the 13 filters together, corresponding to an air sampling of 13 weeks. The other system allows circulating 10⁵ m³ of air per week through a PTI type G3 polypropylene filter (500 x 500 mm and folded to 50 x 50 mm size and 23 mm thick as test sample).

In the case of water test samples, 1 l of water, evaporated on a polyethylene film folded down to 50 x 30 mm size and 2.5 mm of thickness, was studied for ¹³⁷Cs activity concentration determination. 0.300 l of water were analyzed, after radium radiochemical separation, incorporated into a capsule filled with activated carbon (Herranz et al., 2006) (diameter = 61.8 mm and thickness = 15.1 mm) to assess activity concentrations of radium isotopes. Iodine is separated from 2.5 l of water following a precipitation method (Standard Methods, 2017) and the final test sample is put on a plain cellulose nitrate membrane filter (40 mm effective diameter and 0.1 mm thick) to obtain ¹³¹I activity concentration in water samples.

For the analysis of ¹³¹I activity concentration in milk, the test sample is the same plain cellulose nitrate membrane filter as that used for the measurement of radioiodine in water, in this case coming from the iodine separation from 2 l of milk (EPA, 1984).

In the case of soil test samples, soil in a 120 ml beaker (diameter = 71.4 mm and thickness = 30 mm) was analyzed.

3. Method

3.1. Test sample measurement and detector calibration

The analysis of samples was carried out by placing the test samples centered on top of the scintillator. There is almost no distance between sources and the detector surface as the bottom thickness of the polystyrene container of test samples is just 1 mm. Different counting times, from a minimum of one day, were selected to study the minimum time necessary to achieve the detection limits required for those types of

samples in radiological environmental monitoring.

Regarding the quantitative activity concentration measurements, ^{131}I and ^{137}Cs were analyzed in air samples, ^{137}Cs in soil and water samples, ^{131}I in milk, water and radioiodine filters and radium isotopes in water samples.

The experimental energy and efficiency calibration of the detector was done with a set of calibration sources for the different geometrical setups of the test samples. The calibration sources were prepared from a standardized certified solution containing 10 radionuclides, with 12 different gamma-ray energies ranging from 60 to 1836 keV (see Table 1). The calibration sources were prepared with the same procedure as that used for samples, with their geometrical (size and shape) and physical characteristics, so eight different calibration sources were obtained. In all of the cases, it was that the source areas were larger than that corresponding to the circular area of detector. Counting times for the calibration sources were selected according to their activities so that statistical uncertainties of the photopeak areas would have a minor contribution to that of the final result. Consequently, photopeak areas contain at least 10^5 counts for every radionuclide.

In Table 1, a list of radionuclides used for the calibration of the detector together with their emission energies and FWHM recorded with a 1.5" x 1.5" CeBr₃ detector and with a conventional 40% coaxial HPGe detector is shown. It can be seen that in the case of the CeBr₃ detector, the FWHM is around 20 times larger than that of a 40% HPGe detector. The energy resolutions obtained are similar to those reported by other authors with detectors using the same scintillator (García-Toraño et al., 2016; Guss et al., 2009).

The photopeaks used in the efficiency calibration are sufficiently separated in the calibration spectra to allow their location and quantification, despite the FWHM values obtained (Table 1) being rather large as compared to those typically found with HPGe detectors.

Photopeak efficiencies obtained in these calibrations (see Table 2) are rather similar to those found for a 40% relative efficiency coaxial HPGe detector from medium to high energies (see Fig. 1 as an example). At energies lower than 125 keV, this CeBr₃ detector shows higher efficiencies than those found in the case of the semiconductor detector. This last effect must be related with the different windows of both detectors. Anyhow, this effect, which depends on the particular design of the detector, does not affect the gamma-ray spectrometry of the radioisotopes studied in this work as their gamma-ray lines are above 125 keV.

Background measurements were performed by preparing blank samples using the same procedures as for the test samples (see Table 3). It should be pointed out that CeBr₃ scintillation detectors do not suffer from the intrinsic background typical of other detectors like LaCl₃:Ce or LaBr₃:Ce. Background counting time is the same one used for test samples: 1 day. In those background measurements, ^{40}K , ^{214}Pb and ^{214}Bi photopeaks were found with counting rates ranging from 0.012 to 0.018, 0.0081 to 0.0088 and 0.0051 to 0.00779 s⁻¹, respectively. Comparing these values with those measured by using standard coaxial

Table 2

Experimental peak efficiencies measured for the cellulose nitrate membrane filter calibration source of the CeBr₃ detector. The filter is positioned centrally on top of the CeBr₃ detector.

Radionuclide	Efficiency in %	Relative standard uncertainty in %
^{241}Am	15.22	1.57
^{109}Cd	22.02	1.27
^{57}Co	16.04	1.17
^{139}Ce	12.25	1.17
^{203}Hg	9.06	1.23
^{113}Sn	7.95	1.17
^{85}Sr	7.02	1.17
^{137}Cs	5.96	1.17
^{88}Y	4.46	1.17
^{60}Co	3.08	1.17
^{60}Co	2.46	1.17
^{88}Y	1.18	1.17

HPGe detectors with 40% and 60% relative efficiency shielded with 10 cm lead (see Table 3), we find that the 1.5"x1.5" CeBr₃ detector would have a background count rate located between both of them. Contribution of ^{40}K , probably coming from the PMT (Knoll, 2010), is also observed in the higher background values measured by the CeBr₃ detector. As procedures for Ra in water and I in air use activated carbon for its measurement, background count rates from their blank samples show higher values under the ^{40}K photopeak, due to the intrinsic activity of activated carbon in this isotope. Radon progeny background count rates are due to the air volume around detectors inside the shielding, as values from the different detectors are rather similar and they are also similar to the count rates observed in our laboratory when measuring just background, with no blank samples, in detectors with steel shielding.

3.2. Activity concentration, uncertainty and detection limit calculations

Obtained spectra were processed by means of EG&G ORTEC Gamma Vision 7.01 commercial software and, for each measurement, the activity concentration, the uncertainty and the detection limits were obtained. Combined uncertainties were obtained following GUM (ISO, 2008), considering the following sources of uncertainty: sample mass or volume, radiochemical separation yield (cases of Ra and I isolation), calibration (standard certificate, preparation, counting and calibration fitting for gamma-ray spectrometry) and sample and background counting. Decision thresholds and detection limits were obtained following ISO 11929-1 standard (ISO 11929, 2019).

3.3. Precision and accuracy

In order to check the validity of the gamma-ray spectrometry measurements made with this type of detector, several filter, water and soil reference materials from different proficiency tests organized over a number of years by the International Atomic Energy Agency (IAEA) were analyzed and the accuracy and precision were checked for reliability.

The same statistical analyses and criteria that the IAEA would have applied to those results if they had been reported in the framework of those proficiency tests are applied to evaluate those results. These analyses and criteria are the following:

According to the ISO standard 13528 (ISO, 2015), the accuracy was obtained by comparing the relative bias to the maximum acceptable relative bias (MARB).

The relative bias is calculated following equation (1),

$$B = \frac{|A - A_{ref}|}{A_{ref}} \times 100 \quad (1)$$

where A is the measured activity concentration and A_{ref} is the reference activity concentration value.

The MARB values are provided by the organizers, for each radionuclide and for each exercise, considering the method to be used to determine the radionuclide, its activity level in the sample and, in general, the complexity of the task. If $B \leq \text{MARB}$ the result will be accepted for accuracy.

To obtain the precision of the method, the expanded relative combined uncertainty is calculated following equation (2):

$$P = \sqrt{\left(\frac{u(A)}{A}\right)^2 + \left(\frac{u(A_{ref})}{A_{ref}}\right)^2} \quad (2)$$

where $u(A)$ and $u(A_{ref})$ are the combined standard uncertainties of the measured activity concentration A and the reference activity concentration value, A_{ref} , respectively.

If both $P \leq \text{MARB}$ and $B \leq 2.58 \cdot P$, results are accepted for the precision.

Results obtained are shown in the next section.

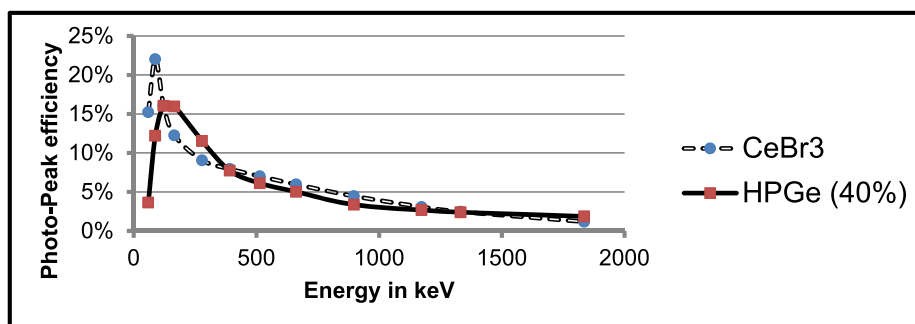


Fig. 1. Comparison of photopeak efficiency of the CeBr₃ and 40% coaxial HPGe detectors obtained for the cellulose nitrate membrane filter calibration source, positioned centrally on top of the detectors, respectively.

Table 3

Background count rates, in 10^{-3} s^{-1} , obtained under the ^{40}K (1460 keV), ^{214}Pb (352 keV) and ^{214}Bi (609 keV) peaks from 1 day counting time of blank samples of soil and blank samples used for the procedures of analysis of radium isotopes in water, ^{131}I in air (through activated charcoal filter for radioiodine collection) and in water, using CeBr₃, 40% HPGe and 60% HPGe detectors.

Blanks/Detector	^{40}K				^{214}Pb				^{214}Bi			
	Soil	Ra in water	I in air	I in water	Soil	Ra in water	I in air	I in water	Soil	Ra in water	I in air	I in water
CeBr ₃	12.0	16.1	17.6	11.6	8.06	8.32	8.22	8.81	5.09	7.72	7.13	6.83
40%HPGe	2.24	9.65	11.3	6.52	5.91	3.63	5.17	8.48	5.06	2.69	4.76	9.11
60%HPGe	4.85	17.2	18.1	7.37	7.82	9.79	8.23	8.96	8.01	9.18	9.06	8.16

4. Results and discussion

Two challenges arise when using a gamma-ray spectrometer for environmental monitoring. The first one is related to the ability of such detectors to properly separate the peaks recorded, coming from the radionuclides of interest, from other interfering peaks, i.e. its resolution. The second one is related to the ability of those detectors to obtain the regulatory detection limits, in our case defined by the CSN and EURATOM. Taking into account the experimental results obtained in this work, the achievement of both challenges is analyzed in this section.

Considering the energy-resolution values obtained for CeBr₃ detectors, it can be concluded that when photopeaks, other than those of the isotope analyzed, appear in the spectrum coming from the background or from the sample itself, with energies close to them, activity assessment can be very difficult as resolving the contribution of the isotope analyzed in the energy peak observed can be rather complex. Another effect of this peak overlapping is the enlargement of its detection limit, as the number of counts under the peak grows with the presence of those photopeaks nearby.

The scope of these effects in the cases analyzed is as follows:

In the case of ^{131}I , the presence of the 352 keV peak from ^{214}Pb in the background complicates the assessment of ^{131}I activity as its main energy peak is at 364 keV, having a FWHM of around 20 keV, and it raises the value of the detection limit of ^{131}I .

In the case of the analysis of radium isotopes, the ^{224}Ra activity is impossible to assess from its gamma-ray peak (241 keV) as it overlaps with those from ^{214}Pb (242 keV) and from ^{212}Pb (239 keV), potentially present in the sample as daughters of ^{226}Ra and ^{228}Ra , respectively. The measurement of the ^{224}Ra activity concentration through the gamma-ray line of its daughter, ^{212}Pb , four days after the Ra-Pb separation when both ^{224}Ra and ^{212}Pb reach a transient equilibrium (Herranz et al., 2006), is also really difficult because of the proximity of those two lines from ^{214}Pb and ^{212}Pb . The ^{226}Ra activity can be assessed by the 609 keV peak of ^{214}Bi , when radioactive equilibrium is achieved, but not from the 352 keV peak of ^{214}Pb , because it is altered by the presence of the 338 keV peak of ^{228}Ac , a ^{228}Ra daughter. The interference of these two gamma-ray energies depends on the ^{226}Ra to ^{228}Ra ratio in the sample. The ^{228}Ra activity can be assessed by the ^{228}Ac activity from its main peak at 911 keV, as it does not have nearby photopeaks.

Fortunately, the ^{137}Cs main peak at 662 keV is clearly separated from the main photopeak of ^{214}Bi at 609 keV in the spectra taken with the CeBr₃ detector.

From the point of view of the detection limits (DL) obtained, the following conclusions can be drawn, looking at the results of radioactivity measurements of several samples (see Table 4).

Once the activity concentrations in the different samples are measured, it can be concluded that, in the case of air filter cartridges using one day as counting time, the detection limit for ^{131}I is between (1.1 and 1.2) 10^{-3} Bq/m^3 . This value is slightly above the detection limit for radioiodine of $1.0 \cdot 10^{-3} \text{ Bq/m}^3$ established by the CSN, (CSN, 1993). However, the use of a counting time of two days for the measurement would allow the achievement of the detection limit required, as, in this case, the detection limit found is $6.5 \cdot 10^{-4} \text{ Bq/m}^3$, which is well below the regulatory value.

In the case of cellulose nitrate air filters and the polypropylene filter, the measurement of the activity concentration of ^{131}I with a counting time of one day achieves a detection limit of around $6 \cdot 10^{-4} \text{ Bq/m}^3$ and $6 \cdot 10^{-6} \text{ Bq/m}^3$, respectively, which is below the required value of $1.0 \cdot 10^{-3} \text{ Bq/m}^3$ established by the CSN, (CSN, 1993).

The measured detection limits of the activity concentration of ^{131}I in air filters are well below the requirement for the detection limit of $2 \cdot 10^{-2} \text{ Bq/m}^3$ for the airborne discharged activity of ^{131}I from nuclear power reactors and reprocessing plants stated by the European Commission Recommendation (Commission Recommendation, 2004).

The measurement, during one day of counting, of the ^{137}Cs activity concentration of air samples collected on cellulose nitrate air filters, on the 13 cellulose nitrate air filters and on the polypropylene filters gives detection limits of $1.5 \cdot 10^{-4} \text{ Bq/m}^3$, $3.1 \cdot 10^{-5} \text{ Bq/m}^3$ and $3.8 \cdot 10^{-6} \text{ Bq/m}^3$, respectively, with the latter two below the required value of $7.4 \cdot 10^{-5} \text{ Bq/m}^3$ established by the CSN, (CSN, 1993). Therefore, a counting time of one day employing this CeBr₃ detector is enough to achieve the requirements for ^{137}Cs in air monitoring when a large volume of air has circulated through the filters. In the case of one cellulose nitrate air filter used for the sampling of 300 m^3 of air in one week, two days of counting time are needed to achieve that required limit. It can also be seen that the detection limits found are also well below the requirement for a detection limit of $3 \cdot 10^{-2} \text{ Bq/m}^3$ in the case of the airborne discharged activity measurement of ^{137}Cs from nuclear power reactors and

Table 4

Detection limits (DL) obtained from the activity concentration measurement of different samples by the equipment under test and by a 40% HPGe detector for comparative purposes. Counting times (T) and sample volumes (V) or sample masses (m), in case of (a) and (b) tables, respectively, are shown as well as the regulatory detection limits defined by the CSN and by EURATOM (for ^{226}Ra and ^{228}Ra).

		CeBr ₃			HPGe			Requirements
		T in h	DL in Bq/m ³	V in m ³	T in h	DL in Bq/m ³	V in m ³	DL in Bq/m ³
Air filter cartridge	^{131}I	48	6.50E-04	3.06E+02	24	3.60E-04	3.06E+02	1.00E-03
Cellulose filter	^{131}I	24	5.80E-04	3.02E+02	24	7.90E-05	3.02E+02	1.00E-03
	^{137}Cs	48	7.38E-05	3.02E+02	24	3.10E-05	3.02E+02	7.40E-05
13 Cellulose nitrate filter	^{137}Cs	24	3.00E-05	3.89E+03	24	4.50E-06	3.89E+03	7.40E-05
Polypropylene filter	^{131}I	24	5.80E-06	8.48E+04	24	4.00E-06	8.42E+04	1.00E-03
	^{137}Cs	24	3.80E-06	8.48E+04	24	4.00E-07	8.42E+04	7.40E-05
Water	^{137}Cs	24	1.40E+02	1.00E-03	24	2.20E+01	1.00E-03	2.00E+02
	^{131}I	24	4.40E+01	2.50E-03	24	1.80E+01	2.50E-03	5.00E+01
	^{226}Ra	24	2.21E+03 (from ^{214}Bi)	3.00E-04	24	3.70E+02 (from ^{214}Pb)	3.00E-04	4.00E+01
	^{228}Ra	24	5.23E+03	3.00E-04	24	9.90E+02	3.00E-04	2.00E+01
Milk	^{131}I	24	3.10E+01	3.50E-03	24	2.30E+01	2.00E-03	3.50E+01
(a)								
		CeBr ₃			HPGe			Requirements
		T in h	DL in Bq/kg	m in kg	T in h	DL in Bq/kg	m in kg	DL in Bq/kg
Soil	^{137}Cs	168	1.03E+00	1.20E-01	168	1.00E+00	1.20E-01	1.00E+00
(b)								

reprocessing plants stated by the European Commission Recommendation (Commission Recommendation, 2004).

For water samples evaporated on a polyethylene film, the detection limit obtained for the measurement of the activity concentration of ^{137}Cs is around 140 Bq/m³ which is also below the required value of 200 Bq/m³ (CSN, 1993) and of 500 Bq/m³, which is the maximum detection limit specified by the Council of the European Union for the activity concentration of ^{137}Cs (Council Directive, 2013) in the requirements for the protection of the health of the general public with regard to radioactive substances in water, intended for human consumption. Although these European detection limits are not intended for environmental surveillance, they are sometimes compulsory for this purpose. Therefore, one day of measurement with this type of detector and a 1 L sample is sufficient to achieve the requirements of detection limits for water monitoring purposes.

If iodine is separated, at least 2.5 L of water are needed to achieve the ^{131}I required detection limit of 50 Bq/m³ for drinking water (CSN, 1993), with one day as counting time. However, only 250 ml of water would be needed to achieve the maximum detection limit for the activity concentration of ^{131}I in drinking water of 500 Bq/m³, specified by the Council of the European Union (Council Directive, 2013).

In the case of the activity concentration determination of ^{131}I in milk, using the same counting time, 3.5 L are needed so that the detection limit found is below the regulatory value of 35 Bq/m³ of the CSN.

The Food and Agriculture Organization of the United Nations together with the World Health Organization published (last modification in 2019) the Codex Alimentarius (FAO-WHO, 1995) in 1995 in which the guidelines levels for activity concentrations of certain radionuclides to be applied to commodities moving in international trade are established. For ^{131}I , guideline levels for activity concentrations are the same for infant and general foods: 100 Bq/kg. On the other hand, the usual practice in environmental monitoring laboratories requires detection limits being 10 times lower than the guideline values (IAEA, 1989), so that a detection limit for ^{131}I activity concentration would have to be lower than 10 Bq/kg, that is, around 10000 Bq/m³ in the case of milk. So, for the case under study, only 20 ml of milk would be needed to reach this value.

As regards soil samples, having used a 120 ml cylindrical beaker on top of the detector, a larger counting time must be used. Around 7 days are needed to get a detection limit for the activity concentration of ^{137}Cs

below the regulatory value of 1 Bq/kg of the CSN.

For the activity concentration of radium isotopes in water samples there is no detection limit established by the Spanish Nuclear Regulatory Authority for radiological environmental monitoring. However, we can look at the specified limits of detection that should be achieved by the methods of analysis of some radionuclides in the analysis of drinking water established by EURATOM, (Council Directive, 2013), even though they are not intended for environmental surveillance. In the case of ^{226}Ra and ^{228}Ra , they are 40 and 20 Bq/m³ respectively (see Table 4). In the case of ^{226}Ra , a sample of at least 10 L and a counting time of 3 days would be needed to reach the detection limit required, but in the case of ^{228}Ra , an excessive counting time and volume of water are necessary to achieve the very low detection limit of 20 Bq/m³.

Clearly, all achieved detection limits of the activity concentration measurement of different samples by the equipment under test are well below those levels established by the IAEA for exclusion, exemption and clearance (IAEA, 2004).

In Table 4, these results are summarized and compared with those obtained when a 40% HPGe detector is used for measuring the activity concentration of the same samples. As it can be seen in this table, for most cases, the same conditions are acceptable concerning both detectors to achieve regulatory detection limits. Only in the case of radioiodine some enlargement must be made regarding the sample volume or counting time to achieve the corresponding regulatory detection limits. In the case of air filter cartridges, more measuring time is needed with this CeBr₃ detector, and in the case of milk, the detection limit required can be met by increasing the quantity of milk, as an easier option, rather than increasing the counting time.

Finally, the results of the measurements carried out by using this CeBr₃ detector compared with reference values in order to test precision and accuracy are shown in Table 5.

Taking into account the values shown in Table 5 and the acceptance criteria explained in section 3.3, it can be concluded that all the results would have been accepted for both precision and accuracy if they had been provided as laboratory results in the framework of the respective proficiency tests.

Thus, the use of this CeBr₃ scintillation detector applied for in lab gamma-ray spectrometry for some interesting radionuclides activity concentration assessment in the field of the radiological environmental monitoring is very reliable.

Table 5

Results of proficiency test. All reference samples come from different proficiency tests organized by IAEA in different years. MARB values are taken from the respective proficiency tests evaluations. Radium isotopes are evaluated following a radiochemical separation process.

Year	Sample	Radionuclide	<i>Aref</i> in Bq/kg	u(<i>Aref</i>) in Bq/kg	<i>A</i> in Bq/kg	u(<i>A</i>) in Bq/kg	<i>B</i> in %	<i>P</i> in %	2.58- <i>P</i> in %	<i>MARB</i> in %
2015	Water	¹³⁷ Cs	30.10	0.90	29.60	0.60	1.66	3.61	9.32	15
2015	Soil	¹³⁷ Cs	715.00	30.0	637.0	39.0	10.91	7.42	19.15	15
2019	Air filter	¹³⁷ Cs	13.25	0.40	12.96	0.67	2.19	5.99	15.45	20
2019	Water	²²⁸ Ra	22.08	1.00	22.30	1.70	1.00	8.87	22.88	20
2019	Water	²²⁶ Ra	7.50	0.25	9.22	0.88	22.93	10.11	26.08	25
Year	Sample	Radionuclide	<i>Aref</i> in Bq	u(<i>Aref</i>) in Bq	<i>A</i> in Bq	u(<i>A</i>) in Bq	<i>B</i> in %	<i>P</i> in %	2.58- <i>P</i> in %	<i>MARB</i> in %
2019	Air filter	¹³⁷ Cs	13.25	0.40	12.96	0.67	2.19	5.99	15.45	20

5. Conclusions

The use of CeBr₃ scintillation detectors for in-lab environmental radiological monitoring has great advantages related to its low price and the low maintenance cost needed. However, its energy resolution is a crucial factor in gamma-ray spectrometry. Its medium resolution could be acceptable to perform the analysis of the different emission lines of important radioisotopes whose activity concentrations are assessed in radiological environmental monitoring, like ¹³¹I and ¹³⁷Cs. However, photopeaks from the sample or the background located near the photopeak under analysis make it difficult to perform spectral analysis effectively and enlarge detection limits of some radionuclides.

In this paper, the evaluation of the activity concentrations of ¹³¹I, ¹³⁷Cs and radium isotopes in different measurement geometries is performed in spite of the fact that some peaks overlap complicating their proper assessment.

It is shown how the use of one day counting time, as commonly used with HPGe detectors, allows the attainment of the detection limits required by the Nuclear Safety Council of Spain and those of the European Council directive for radioactive substances in drinking water. However, in some cases, the use of larger counting times or larger sample volumes, if possible, are required to achieve those detection limits. Of course, the achieved limits are well below those levels established by the IAEA for exclusion, exemption and clearance.

The validity of the results obtained from different samples and radionuclides has been proven by comparison with reference values for samples from proficiency tests.

Consequently, this 1.5" x 1.5" CeBr₃ scintillation detector that operates at room temperature and which is considerably cheaper than traditional HPGe detectors, could be considered as a good option for simple routine tasks associated with in lab radiological environmental monitoring as it can perform excellently in assessing selected radionuclides that must be continuously monitored in the environment.

As a result, this CeBr₃ scintillation detector makes it possible to significantly reduce costs associated with radiological environmental monitoring. The cost reduction is threefold: the detector itself, the maintenance cost are reduced, as no cooling is required, and, due to the smaller dimensions of the detector, the shielding can be designed much smaller as well, reducing costs for shielding materials and its installation.

Author statement

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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