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Advances in Instrumentation and Methods for Environmental Radioisotope Monitoring

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Radiation monitoring apparatus and methods are classified by main operation characteristics – minimal detectable activity (MDA) and the capability to separately identify nuclides from a mixture of several radionuclides. The lowest MDA per unit volume is achieved by a large-size device (large sample and scintillation crystal) using high-energy γ-photons for the identification of nuclides; MDA strongly depends on the size and effective atomic number of the crystal. If the examined sample contains a number, K, of nuclides, their separate identification without limitation of K can be carried out by the fitting method (the amplitude spectrum of measured signals is expanded over the spectra of separate nuclides). In cases where MDA per unit volume is less important, nuclides can be identified by β-particles and low-energy γ-photons with the use of a compact device – phoswich detector with two scintillation layers strongly differing in thickness and decay time.

KEYWORDS: *environmental radioisotope monitoring,* ***radionuclide, minimal detectable activity, detection of low activities.***

I. Introduction

The detection and identification of low-active radionuclide pollutions is, as a rule, carried out with the use of a scintillation detector systems and is based on the measurement of the energy spectrum of signals produced by the substance under investigation (see, e.g. Refs. [1 - 8]). Along with this, measurements of nuclides can be carried out by the inductively coupled plasma mass-spectrometry [9]. Below we will restrict ourselves to scintillation detectors.

The efficiency of the detection method is characterized by the minimal activity (MDA) per unit volume of the substance which can be reliably registered on the radioactive background. MDA is determined by the statistical fluctuations of the background activity: the number of reliably registered signals must exceed the dispersion of the background counts.

There exist different types of apparatus for detecting low-active nuclides. Below we will restrict ourselves to two most widespread types:

(1) A setup, using high-energy γ-emission of nuclides for their identification, is based on a large scintillation crystal. A large sample of the examined substance is placed into a Marinelli beaker with a cavity for the crystal. The apparatus is placed inside a heavy protection shell to diminish background activity. Such setup, at the expense of large sizes and weight, provides a low level of MDA. The latter depends essentially on the effective atomic number, Z_{eff}, of the scintillator as well as on its size.

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(2) A setup, that uses short-range particles (in particular, high-energy β-particles and low-energy γ-photons) for detecting and identifying nuclides, contains both a sample and scintillator in the form of plates. The use of two different scintillation layers (phoswich detector) provides additional possibility for separating nuclides.

The apparatus of the former type, using high-energy γ-photons, is considered in Sec. II with especial attention to the effect of the scintillator atomic number Z_{eff} on MDA as well as to the optimal way of processing signals.

In Sec. III, the detection apparatus on the base of phoswich detectors is considered and the MDA levels, related to the apparatus of two types, are compared.

II. Apparatus and methods for detecting nuclides by high-energy γ-photons with the use of a large scintillation crystal and Marinelli beaker

A large path length of high-energy γ-photons makes it expedient to use a large scintillation crystal with a sufficient Z_{eff} and a sample even larger (the stopping power of the sample is usually much less than that of the crystal substance). Such setup consists of a Marinelli beaker that has a cavity for placing the crystal inside the sample. The apparatus is placed inside a heavy protection shell to suppress the background activity.

There exists a large variety of Marinelli beakers. As an example, we will consider two versions which differ by the Marinelli beaker volume of 1000 or 500 cm^3 and by the scintillation crystal size of ∅ 63 × 63 or ∅ 40 × 40, respectively.

The radionuclides, contained in the sample, are detected and identified by the amplitude spectrum of the

photomultiplier (PMT) signals (PMT is used as a photoreceiver to enhance the sensitivity of the setup). To identify the nuclides, the measured amplitude spectrum can be processed by two methods considered below.

1. Detection and identification of nuclides via counting γ-photons within a chosen line of complete absorption

The counting method is most widespread (see, e.g., [4 - 8]) and consists in what follows. A radionuclide sample contains a set of nuclides. Each nuclide is detected by using its strongest radiation line, E_0, not overlapping with the rest of the lines of the set. The corresponding detection efficiency D is defined as the ratio of the number of γ-photons with energy E_0 completely absorbed in the crystal to the total number of emitted γ-photons with the energy E_0. MDA per unit volume, corresponding to the nuclide detection probability of 95%, has the form [1)]

$$\frac{\mathrm{MDA}}{V} = \frac{1.18}{DPV}\sqrt{\frac{A_{bac}}{t}} = \frac{1.18}{\eta V}\sqrt{\frac{A_{bac}}{t}} \qquad (1)$$

Here V is the nuclide sample volume, A_{bac} denotes background activity (the number of decays per second within the energy window around the chosen line E_0), t is the exposition time, P stands for the portion of the chosen emission line in the total activity of the nuclide. The detection device is characterized by a sensitivity $\eta = PD$.

Detection efficiency D plays a crucial role in the formation of MDA. D depends mainly on the effective

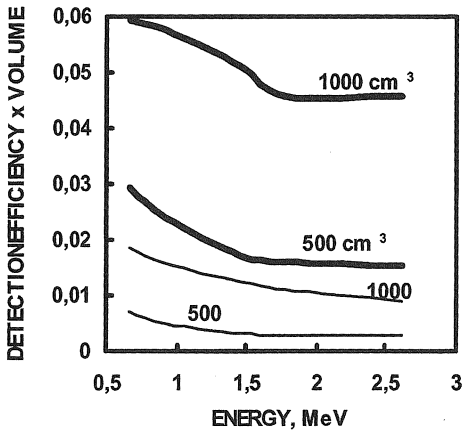

Fig. 1. Detection efficiency D, muliplied by sample volume V, as a function of γ-photon energy (DV mainly determines MDA per unit volume). Thin lines relate to NaI:Tl and thick lines to BGO, sample volume is indicated in the figure.

atomic number, Z_{eff}, and the size of the scintillation crystal. For a given crystal, MDA/V depends also on the sample volume but more weakly than in proportion to $1/V$ since D decreases with an increase of the sample size. These dependences are important for the design of the device; they were partially calculated in Refs. [4 – 8] and, in more detail, in [10]. Some calculation results, obtained in Ref.[10], are presented in **Tables 1 and 2** for four most widespread nuclides which cover the spectral interval 0.06 to 2.6 MeV (the nuclides Ra-226 and Th-232 are presented in the tables by two lines suitable for identification). **Tables 1** and **2** are related to a water sample in Marinelli beakers of the volumes V=1 and 0.5 liter and scintillation crystals with sizes of ∅ 63 × 63 or ∅ 40 × 40, respectively (in all cases, exposition time t=1 hour). The MDA values, given in the last column of the tables, agree with the corresponding experimental data with the accuracy of 15 % (MDA is measured experimentally with the same accuracy).

The values of MDA/V, presented in the tables, vary in a broad interval from 0.8 to 50 decays per second and liter. In accordance with Eq. (1), MDA per unit volume as a function of energy is mainly dictated by the detection efficiency D shown in **Fig. 1**. As seen from the figure, DV increases about 3 times when changing from NaI:Tl to a heavier scintillator BGO. Nearly the same effect accompanies an increase of the volume of the sample by the factor of 2 and the corresponding increase of the scintillation crystal size. The enhancement of Z_{eff} when changing from NaI:Tl to BGO result in a significant effect: the exposition time, required to achieve the same MDA, is reduced about 6 times.

Irrespective of the above considerations of detection efficiency, there is a general tendency to a decrease of MDA with increasing energy due to a strong decreasing energy dependence of background activity (this can be seen from **Tables 1** and **2**).

The MDA values, presented in **Tables 1** and **2**, were obtained under an assumption that the examined sample contains only one nuclide. In the case of a mixture of nuclides, their identification is hampered by a partial overlap of their complete absorption lines present in the amplitude spectrum. One can get a notion on such an overlap from **Fig.2**, where the amplitude spectra of four widespread nuclides are shown. If the sample contains a mixture of nuclides, it is necessary to separate the complete absorption lines belonging to different nuclides, which requires an additional analysis and entails an enhancement of MDA.

Table 1. Detection efficiency, D, and MDA related to 1 liter of the examined substance (sample volume V=1 liter, crystal size ∅ 63×63 mm, exposition time t=1 hour)

Scintil-lation crystal	Nuc-lide	Chosen line energy (MeV)	P	DV (liter)	A_{bac} (decays per sec)	MDA (decays per sec & liter)
NaI:Tl	^{226}Ra	0.609	0.495	0.0283	1.40	2.81
BGO				0.0584	1.72	1.51
NaI:Tl	^{137}Cs	0.662	0.85	0.0292	1.30	1.53
BGO				0.0594	1.60	0.83
NaI:Tl	^{232}Th	0.930	0.45	0.0239	0.80	2.76
BGO				0.0575	0.98	1.27
NaI:Tl	^{40}K	1.460	0.105	0.0164	0.48	13.4
BGO				0.0500	0.59	4.88
NaI:Tl	^{226}Ra	1.764	0.17	0.0159	0.20	5.52
BGO				0.0456	0.25	2.13
NaI:Tl	^{232}Th	2.614	0.36	0.0155	0.15	2.30
BGO				0.0456	0.18	0.86

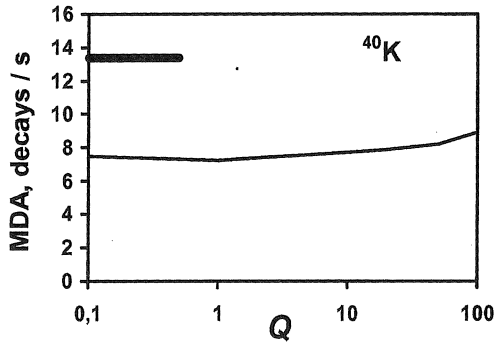

Fig. 3. The fitting method results [13]: MDA for ^{40}K (E_γ= 1.46 MeV) versus activities of other nuclides (Q is the ratio of activities defined by Eq. (10)) . Short bold line shows the MDA value obtained by the counting method under the same conditions but for a single nuclide.

Of most interest is the dependence of MDA(k), defined for a given kth nuclide, on the activities of the rest of nuclides contained in the sample. To investigate this dependence, the activities of the rest of nuclides A_l ($l \neq k$) were taken equal to each other and the ratio

$$Q = A_l / A_k \quad (k \neq l) \qquad (3)$$

was varied.

Fig. 3 presents some results[13] related to the 1 liter Marinelli beaker and NaI:Tl crystal with the sizes ∅ 63×63 mm (exposition time t=1 hour). **Figure 3** shows the dependence of MDA of a chosen kth nuclide on the ratio Q. The short horizontal line indicates the corresponding MDA value obtained by the counting method for a sample with a single nuclide (Q=0).

It can be seen from **Fig. 3**, first, that the MDA, found by the fitting method, is less than the corresponding MDA obtained by the counting method (this was proved for ^{40}K, ^{232}Th, ^{226}Ra, ^{137}Cs[13]). Second, within a broad range of Q, the fitting method gives the MDA of a given nuclide practically insensitive to the content of the rest of nuclides despite the overlapping of their spectra. Note that at a large Q the advantage of the fitting method as compared to the counting method becomes significant: in the presence of other nuclides, the efficiency of the counting method must be noticeably lower because of overlapping of the complete absorption lines.

On the other hand, the application of the fitting method requires the accurate measurement of the basis spectra and the corresponding sensitivities for all nuclides.

III. Identification of nuclides by short-range particles with the use of phoswich detectors. Comparison with a large-size device by MDA/V.

Along with long-range high-energy γ-photons, short-range particles emitted by nuclides provide an additional possibility of their identification. However, it is obvious a priori that MDA per unit volume must be generally large because of a small useful part of the sample volume restricted by a short particle path length l (α-particles with a very small l hardly are suitable for this application).

A most efficient detection setup using short-range particles is a so-called phoswich detector consisting of two scintillation layers strongly differing in thickness ($h_1 << h_2$) and in the scintillation decay time τ. The layer thicknesses are matched to strongly differing path lengths of different-type particles, so that each of them is absorbed mainly in the corresponding scintillation layer. The PMT signals from the layers are discriminated by τ.

Phoswich detectors can be fabricated in two ways:

(1) The liquid phase epitaxy method[14] provides the possibility to grow a thin scintillation film of an arbitrary thickness $h_1 \geq 1$ μm on a scintillation substrate of the thickness $h_2 >> h_1$.

(2) A thin layer (film) can be deposited onto a thick layer (substrate) by thermal evaporation in vacuum. Let us consider in detail the following example. CsI:Tl film deposited onto a polystyrene substrate[15] (**Fig. 4**). The CsI:Tl polycrystalline layer about 40 μm thick is used for detecting soft γ-photons or X-rays, and the 5 mm thick polystyrene substrate detects high-energy β-radiation. With allowance for the luminescence maximum position of CsI:Tl and polystyrene, 550 and 430 nm, respectively, such phoswich detector is coupled to PMT. The signals from the film and substrate are easily separated by their decay times of about 800 and 10 ns, respectively.

SAMPLE
50 μm thick CsI:Tl for soft γ-rays, τ=800 ns
Polystyrene for detecting β-particles 5 mm thick τ=10 ns
PMT

Fig. 4. An example of phoswich detectors: a thin CsI:Tl film for detecting soft γ-photons or X-rays, deposited onto a polystyrene substrate. The latter serves for detecting high-energy β-particles.

This phoswich detector is used to examine the samples containing the mixture of widespread nuclides (presented in **Table 3**) in order to determine their concentrations. To that end, the PMT signals from the film and substrate are separated by their decay times and processed by a special computer program.

The identification of nuclides by a phoswich detector is to a some degree hampered by a continuous energy distribution of β-particles (emitted simultaneously with neutrino). In view of the absence of narrow lines in the amplitude spectrum of signals produced by β-particles, the counting of

Table 2. Detection efficiency, D, and MDA related to 1 liter of the examined substance (sample volume V=0.5 liter, crystal size ∅ 40×40 mm, exposition time t=1 hour).

Scintil-lation crystal	Nuc-lide	Chosen line energy (MeV)	DV (liter)	A_{bac} (decays per sec.)	MDA (decays per sec. & liter)
NaI:Tl	^{226}Ra	0.609	0.00072	0.80	8.38
BGO			0.00192	0.65	2.82
NaI:Tl	^{137}Cs	0.662	0.00070	0.76	4.92
BGO			0.00185	0.60	1.64
NaI:Tl	^{232}Th	0.930	0.00049	0.47	10.4
BGO			0.00157	0.37	2.86
NaI:Tl	^{40}K	1.460	0.00032	0.25	50.4
BGO			0.00124	0.20	11.5
NaI:Tl	^{226}Ra	1.764	0.00030	0.12	23.0
BGO			0.00112	0.09	5.26
NaI:Tl	^{232}Th	2.614	0.00027	0.09	10.5
BGO			0.00088	0.07	2.8

For a mixture of nuclides, the fitting method of processing signals is more adequate since the entire amplitude spectra of different nuclides are essentially distinguished from each other even under an overlap of some their lines of complete absorption.

2. Detection and identification of nuclides by the fitting method of processing amplitude spectrum γ- photons

The measured spectrum of signals can be processed also in another way using the information contained in the entire spectrum (containing all the lines of complete absorption without excluding overlapping ones and the Compton continuum). This approach uses the basis spectra, related to the samples containing a single nuclide each. The spectrum of registered signals, produced by a sample with a set of nuclides, is approximated by the linear combination of the basis spectra. The coefficients of this expansion, found via its fitting with the measured spectrum, give the content of every nuclide (see, e.g., [11, 12]). This way of nuclide identification will be for brevity referred to as the fitting method.

At first sight, the fitting method is inferior to the counting method in view of a much greater number of background counts registered within the entire spectrum (note that within the Compton continuum the signal-to-background ratio is much less favorable than in the vicinity of a complete absorption line). For this reason, the fitting method practically was not used up to now.

However, as was shown in Ref. [13], the above demerit of the fitting method is overcompensated by a large volume of information derived from the Compton continuum and all complete absorption lines related to each nuclide. The essence of the fitting method consists in what follows.

The sample contains a set of K low-active nuclides. The activity of each nuclide should be derived from the measured energy spectrum of signals from the sample, S_m, i.e. the number of signals registered during the exposition time t within the mth channel of the detection system. The spectrum S_m (subject to statistical fluctuations) includes the activity of the sample and background activity b_m. The basis spectrum $F_{m,k}$ is defined as the normalized number of signals, emitted by the k-th nuclide within the m-th channel, and does not include the background activity. The basis spectra are exactly measured during a long time and are not subject to statistical fluctuations.

The aim of the measurement is to find the total emitted activities of every nuclide, A_k, defined as the mean number of γ-photons emitted by the sample per unit time. For that end, the measured spectrum S_m is approximated through the activities A_k. The approximated spectrum, S_m^{app}, consists of the background activity contribution tb_m and the contribution of the examined nuclides:

$$S_m^{app} = tb_m + t\sum_k F_{mk} H_k A_k$$

where t is exposition time, H_k is the integral sensitivity, i.e. ratio between the total numbers of the detected and emitted γ - photons for the k-th nuclide. Following the standard least square method, the mean square discrepancy between

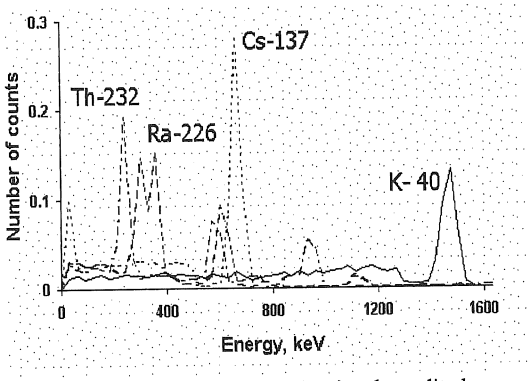

Fig. 2. Basis spectra of the PMT signal amplitudes converted to γ-photon energy [13].

the approximated and measured spectra are minimized with respect to nuclide activities A_k. This results in the set of K equations for the total activities of K nuclides:

$$\sum_k L_{pk} A_k = M_p \qquad (2)$$

with coefficients

$$L_{pk} = t\sum_m F_{mk} F_{mp} H_k H_p\,,\quad M_p = \sum_m (S_m - tb_m) F_{mp} H_p\,.$$

The mean value of the k-th nuclide activity, A_k, obtained from Eq. (2), differs from its exact value because of the statistical fluctuations of the measured spectrum S_m. For the kth nuclide, MDA was defined as the mean value of the activity A_k providing the detection of the kth nuclide with the probability of 95%.

signals should be complemented by an additional analysis of their amplitude spectrum [15]. For that purpose, the fitting method (described in Sec. 2.2) seems more adequate.

However, the counting method can be used to estimate MDA for a phoswich detector and to compare it with that of large-size setup with Marinelli beaker. For such comparison, let us put in (1) the parameter P (related to a nuclide and not to apparatus) equal to unity and t=1 hour in all cases. To characterize MDA for a setup, using β-particles and low-energy γ-photons, we will restrict the consideration to β-particles for which MDA is greater (even a low-energy γ-photon has a large enough path length in a sample with a small Z_{eff}).

MDA for a phoswich detector is mainly determined by the product DV. For the 2π-geometry (**Fig. 4**), D=0.5 within the examined substance layer of the thickness l_β, so that $DV=0.5 l_\beta S_{PMT}$ (l_β is the β-particle path length in the substance and S_{PMT} is the PMT cathode area).Taking into account the isotropic distribution of emitted β-particles over directions, one obtains

$$DV=0.25 l_\beta(E) S_{PMT} \quad \text{for} \quad h \geq l_\beta(E_{max}) \qquad (4)$$

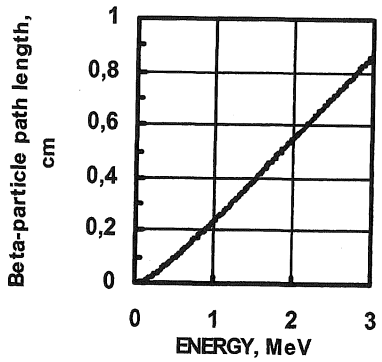

Fig. 5. Energy dependence of the β-particle path length l_β in a substance of the density ρ= 2 g/cm^3 (l_β is proportional to 1/ρ)

where h is layer thickness and E_{max} denotes the maximum β-particle energy within the range considered. It should be emphasized that an increase of h in the region $h \geq l_\beta(E_{max})$ does not change DV; thus, the optimal thickness of the sample is $l_\beta(E_{max})$. The energy dependence of l_β, calculated according to Ref. [16] for the substance density ρ=2 g/cm^3, is shown in **Fig. 5**. Thus, the MDA ratio for the devices considered in Secs. 3 and 2, can be roughly estimated as

$$\frac{(\mathrm{MDA}/V)_{phos}}{(\mathrm{MDA}/V)_{Mari}} \approx \Lambda_{bac} \frac{(DV)_{Mari}}{(DV)_{phos}} = \Lambda_{bac} \frac{4(DV)_{Mari}}{l_\beta S_{PMT}} \qquad (5)$$

where the multiplier Λ_{bac} (near to unity) allows for the background activity differing for the two types of devices.

As an example, let us consider the ratio (5) at a favorable geometry of the devices of both types. For phoswich detector, the areas of the sample and scintillator are determined by the PMT cathode area S_{PMT}=38 cm^2 (with diameter of 7 cm). For the device with Marinelli beaker, the sample volume is 1000 cm^3 and the scintillation crystal measuring ∅ 6.3×6.3 cm has energy resolution of 10% (then Λ_{bac}≈0.7).

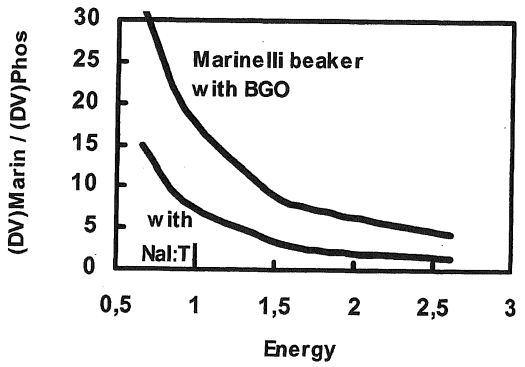

Fig. 6. Comparison of MDA per unit volume for a phoswich detector and a large-size device with the 1 liter Marinelli beaker and the NaI:Tl or BGO scintillation crystal ∅ 63×63 mm.

Table 3. Energy of soft γ-photons (or X-rays), detected by the CsI:Tl film, and β-particles detected by the polystyrene substrate. For β-particles, the energy boundary, E_b, of the β-radiation continuum is indicated (the continuum has maximum at about E_b /3). In the last line of the table, MDA of the setups with phoswich detector and 1 liter Marinelli beaker are compared according to the estimate (5).

Layer of phoswich detector	CsI:Tl film		Polystyrene substrate		
Nuclides	^{239}Pu	^{241}Am	^{90}Sr	^{40}K	^{137}Cs
Soft X-ray or γ-photon energy, keV	14,17, 21 (unresol-ved set)	17 and 59.6	Not used for detection of these nuclides		
Boundary of β-radiation continuum, MeV	Not used for detection of these nuclides		0.550 (^{90}Sr) 2.250 (^{90}Y)	1.3	0.51
$(\mathrm{MDA}/V)_{phos}/(\mathrm{MDA}/V)_{Marinelli}$	8* 8**		15* 27**	27* 45**	110* 130**

*For the device with the 1 liter Marinelli beaker and NaI:Tl crystal.

**For the device with the 1 liter Marinelli beaker and BGO crystal.

IV. Conclusions

The radiation monitoring apparatus and methods should be classified by main operation characteristics – minimal detectable activity (MDA/*V*) and the capability to identify separate nuclides from their mixture.

Such qualitative classification is given in **Table 4**. The lowest MDA is achieved with a large-size device using high-energy γ-photons for identification of nuclides (large sample in a Marinelli beaker and a correspondingly large scintillation crystal). MDA can be additionally reduced about three times via doubling the Marinelli beaker volume or via replacing a NaI:Tl scintillation crystal by a heavier BGO scintillator of the same sizes (see Tables 1 and 2).

In cases where of most importance is a low MDA per unit volume, such large-size apparatus should be used. If, in addition, a mixture of many nuclides is to be examined, the measurement results should be processed by the fitting method. Its application requires the accurate measurement of the basis spectra for the set of nuclides to be examined.

In cases where a low level of MDA is less important, the radionuclide monitoring can be carried out with a compact device on the base of a phoswich detector.

Table 4. Qualitative characteristics of operation capabilities of devices for radiation monitoring

Type of device	Processing method and degree of its complication	MDA range (decays/sec• liter)	Capability of separating nuclides from a mixture
Large sample and large scintillation crystal (1 liter Marinelli beaker) using high- energy γ-photons for identification of nuclides	Counting γ-photons in an energy window near a complete absorption line (requires a minimum information on the amplitude spectrum of signals).	1 to 10	Limited, requires additional analysis
	Fitting a linear combination of basis spectra with the amplitude spectrum of signals measured (requires basis spectra measured with a high accuracy).	0.7 to 7	Practically not limited, completely formalized
Phoswich detector with scintillation layers strongly differing in thickness and decay time	Separate processing of signals from each layer (counting signals and analysis of spectra). Optimal would be the fitting method.	15 to 300	Very good but requires additional analysis

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