RAPID ANALYSIS OF RADIONUCLIDE COMPOSITION (SCREENING) OF LIQUID SAMPLES VIA DECONVOLUTION OF THEIR LS SPECTRA

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ABSTRACT. Liquid scintillation (LS) spectrometry based on an original method for processing initial LS spectra is presented. The algorithm used is based on the modeling of spectra transformed by a convolution into group form by the superposition of individual reference spectra taken from a previously created nuclide library. In this library, for each radionuclide (or radionuclide + DP), there is a set of at least 10 spectra measured at different quenching parameters. The model spectrum is presented as a sum of elemental spectra with weight coefficients (c_j) that are adjusted during the modeling procedure so that the model spectrum will coincide with the sample spectrum to a maximum degree. Due to the development of new approaches for interpreting LS spectra realized in the software RadSpectraDec, the possibilities of this method for implementing a rapid analysis (screening) of samples with unknown radionuclide composition has been considerably extended. The software now allows us to analyze actual complex radionuclide mixtures of environmental or technological origin. The reliability of the proposed procedures has been confirmed by "added-measured" tests, comparative radiochemical analysis, and by analyzing materials with a regulated content of radionuclides (IAEA, RM, and PT). In this work, data are presented that have been obtained with the given approach for analyzing different materials, such as liquid waste, drill water, liquid discharge, potable water, etc. The limitations regarding sample screening operations are considered as well.

INTRODUCTION

In the first phase for examining technological and environmental samples with unknown radionuclide composition, a traditional approach is used: evaluation of the integral activity of alpha and beta emitters and gamma spectrometry. However, measurement of integral alpha/beta activity does not provide any information about the radionuclide composition of the sample. Gamma-ray spectrometry with HPGe detectors is rapid and informative, but for a number of long-lived alpha and beta emitters with high radiotoxicity (such as ^{90}Sr, TRU isotopes, and natural isotopes of Ra, Th, U, ^{210}Pb, ^{210}Po), this method offers very limited information. Therefore, further separation of these radionuclides is performed randomly with certain radiochemical methods, taking into account all possible variants of components.

Liquid scintillation (LS) spectrometry, which has several advantages in comparison with standard approaches, is an alternative method for estimating the radionuclide composition of various samples. LS counters are now widely used for radioisotope assay because they are highly efficient in detecting alpha and beta radiation at a minimum background. The LS counting (LSC) method evaluates the energy of particulate radiation; however, until now the basic problem hindering the use of LS counters as genuine spectrometers was the correct splitting of the multicomponent apparatus spectrum into initial components. Analyzing the multicomponent radionuclide mixtures from environmental or technological systems faces some complications due to both the uncertainty of the mixture composition and the continuity and overlapping of the beta spectra.

As a result of previous investigations (Kashirin et al. 2000a,b, 2003a,b), the possibilities of LSC have been enhanced considerably. After developing new mathematical approaches to decoding complex spectra applied to LSC, realized in the expert software package RadSpectraDec, we were able to immediately obtain information about the composition of the main alpha- and beta-emitting radionuclides, or at least to determine whether they exceeded the control levels for particular radionuclides, a frequent goal of radiation monitoring. The liquid source for counting may be prepared by directly mixing a sample aliquot with LS cocktails, or by preliminary evaporation-concentration. To

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increase the sensitivity of analyzing components in the initial spectrum during the screening examinations, various modifications of coprecipitation and extraction procedures were used.

The developed approach can be applied to all models of modern LS counters, of both linear and logarithmic amplification, and with most types of scintillation cocktails, for a wide range of quenching. This paper presents data obtained by applying this approach for analyzing different materials, such as liquid waste, drill water, liquid leakages, potable water, and others.

MATERIALS AND METHODS

Reagents and Tracers

All chemicals were of reagent grade. We used the LS cocktails Ultima Gold™ AB, Ultima Gold F, Insta-Fluor, and OptiPhase HiSafe 3 (PerkinElmer, USA). All measurements were carried out with low-diffusion 20-mL polyethylene vials.

The following standard radioactive solutions used to create a spectral library were obtained from various institutions: All-Russian Scientific Research Institute of Metrology (Russia), Institute for Physics and Power Engineering (Obninsk, Russia), Joint-Stock Company "Cyclotron" (Obninsk, Russia), and LNP and LNR of Joint Institute of Nuclear Research (Dubna, Russia). ^3H and ^{14}C were purchased from Packard Instrument Company (USA); ^{125}I and ^{32}P were purchased from AEA Technology (UK).

Separation and purification of ^{226}Ra, ^{228}Ra, ^{234}Th, ^{228}Th, ^{224}Ra, ^{210}Pb, ^{210}Bi, ^{210}Po, ^{90}Sr, and ^{90}Y from corresponding standard radioactive solutions were performed using column chromatography with Sr resin (Eichrom Inc., USA) and Dowex 1×4 resin.

Instruments

A Tri-Carb 2550 TR/AB LS analyzer was used in this study. After radiochemical separations, alpha emitters were measured with an Alpha spectrometer model 7401 (Canberra Inc.) equipped with PIPS detectors (effective surface 600 mm^2). Stainless steel disks with electrodeposited radionuclides were used as counting sources. The RadSpectraDec spectra processing software program (Tri-Carb and Alpha versions) was used for identifying radionuclides and calculating their activities.

Radiochemical Methods

For comparative radiochemical analysis, the following techniques were utilized. Brief descriptions are given below.

- Uranium: Extraction with TBP in toluene, purification of organic phase, back-extraction with distilled water, electrodeposition, alpha-ray spectrometry;
- Thorium: Anion-exchange chromatography, LS spectrometry (^{234}Th), electrodeposition, alpha-ray spectrometry;
- Radium: Separation and purification via a series of precipitating procedures (chromate, hydroxide, chromate, and transformation into barium-radium carbonates by metathesis), LSC;
- Polonium: Spontaneous deposition onto polished stainless steel disk from a hydrochloric acid solution in the presence of citric and ascorbic acids, alpha-ray spectrometry;
- Strontium: 1) Separation and purification via a series of precipitating procedures (sulfate, hydroxide, chromate—if necessary, sulfate in presence of EDTA—transformation into stron-

tium carbonate by metathesis); 2) Column sorption chromatography using VS-15 material (Ermakov et al. 2002);
- Plutonium: Anion-exchange chromatography, final purification by extraction with TTA in toluene, electrodeposition, α-ray spectrometry;
- Nickel: Complexation with dimethylglyoxime, extraction with chloroform, back-extraction with hydrochloric acid solution, LS spectrometry;
- Technecium: Anion-exchange chromatography with TEVA resin (Eichrom), LS spectrometry.

Identification of Radionuclides via Modeling Complex LS Spectra

LS spectra generally have a continuous, complicated character, and the constituent parts, correlated to various radionuclides, overlap in a wide energy range. Because of the complexity and variety of the spectra, exposition by a set of some elementary functions is practically impossible. To solve this problem, a novel method was proposed by Malinovsky et al. (2002a) based on modeling of the initial spectra transformed by convolution into group form by the superposition of individual reference spectra taken from the previously created nuclide library.

A model spectrum is created as a linear combination from normalized spectra of various radionuclides:

$$M_i = \sum_{i=1}^{J} c_j M_{i,j}(T) \tag{1}$$

where M_i is the model spectrum, $M_{i,j}(T)$ is the library spectra of each nuclide, and c_j is the proportion of each species. The weight coefficients (c_j) are adjusted so that the model spectrum will coincide with the sample spectrum to a maximum degree. Each spectrum is a function of the quench parameter (T) of the sample. The solution is generated by recursive minimization of a special function representing the difference between a model spectrum and the sample spectrum. Next, the whole spectrum is processed without counting in different energy windows, and the result is an increase in the sensitivity of the measurements.

In order to increase the statistical information, it is necessary to combine a series of neighboring channels. In this process, the initial spectrum is combined into groups, and the algorithm of a grouping depends of the pattern of the spectrometer amplifier.

Preliminary evaluations have shown that for spectrometers with a linear analog-to-digital converter (ADC) (Tri-Carb series), the optimum algorithm for binding apparatus spectra into groups is a quasi-arithmetic progression. The formula of convolution is as follows:

$$G_k = \frac{1+k}{[(1+k)/2]} \sum_{i=N_k}^{n_{k+1}-1} P_i, \, k = 1, 2... K \tag{2}$$

where G_k is the group spectrum and P_i is the initial apparatus spectrum.

For spectrometers with a logarithmic ADC (Quantulus and Guardian from Wallac, Triathler from Hidex) with a detailed low-energy region of the spectrum, a traditional linear combination provides good results (e.g. a combination of 10 channels in a group) (Malinovsky et al. 2002b).

Creation of Spectra Library

The shape of the apparatus spectrum essentially depends on the properties of the prepared liquid counting source. The spectrum of a higher quenched sample is condensed to the low-energy region, sometimes with considerable deformation. For creating the nuclide library, we used direct measurements of each radionuclide spectra. This library of reference spectra is organized for the existing counter and for the appointed characteristics of counting sources by measuring the standard solutions at different quenching parameters. The analytical database (the library of quenched spectra) for each radionuclide was obtained for at least 10 quench levels with CCl_4 as a chemical quench agent. For example, for determining natural decay chains we have the following sets of spectra: ^{238}U, ^{234}Th+^{234}Pa^{m}(equ), ^{232}Th, ^{228}Th, ^{210}Pb, ^{210}Bi, ^{210}Po, ^{235}U, ^{231}Th+^{231}Pa(equ), ^{228}Ra+^{228}Ac(equ), ^{234}U, ^{230}Th, ^{226}Ra, ^{226}Ra+^{222}Rn+^{218}Po+^{214}Pb+^{214}Bi+^{214}Po(equ), ^{224}Ra+^{220}Rn+^{216}Po+^{212}Pb+^{212}Bi+^{212}Po+^{208}Tl(equ). All spectra in the library are normalized and combined into groups of channels (Figure 1).
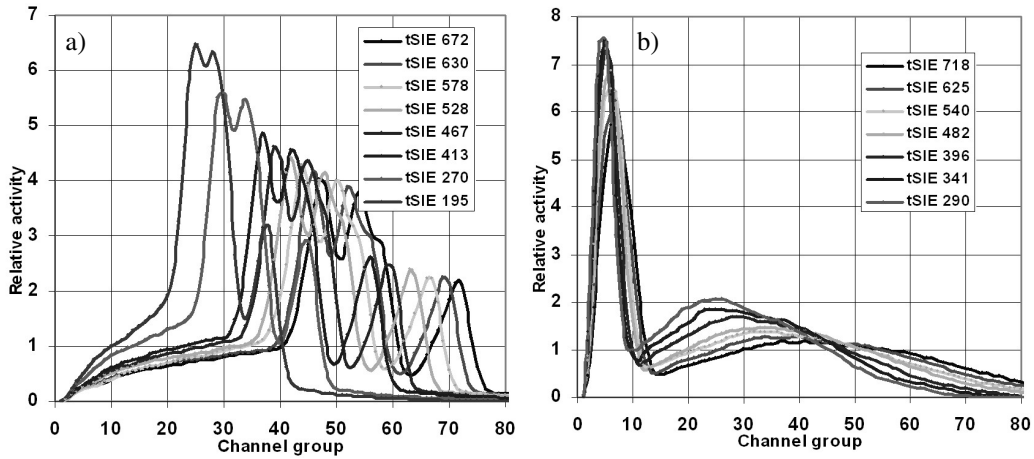

Figure 1 Sets of reference library LS spectra: a) ^{228}Ra+^{228}Ac; b) ^{226}Ra+DP, after converting into group form

While processing the measured spectrum, the shapes of individual library spectra are fitted to the required quenching parameters via an interpolation of two of the neighboring reference spectra. At present, the analytical database (the library of quenched spectra) for the Tri-Carb 2550 TR/AB in SIA Radon consists of about 60 β and α emitters (individual and in equilibrium with the daughter products), which enable one to determine different radionuclides after radiochemical separation. At the same time, we can investigate samples with complex radionuclide composition, such as wastes (solid radioactive waste [SRW] and liquid radioactive waste [LRW]), waters from observation drills, etc., by the screening method after performing simplified sample preparation procedures.

Calculating the Radionuclide Activity

During LS analysis, the activity of a radionuclide (A_j) in a sample may be determined via the parameter c_j, defining a contribution of a radionuclide into a spectrum with the following expression (for $T_{sample} = T_{bkg}$):

$$A_j = \frac{(N - N_{bkg}) \times c_j}{T\varepsilon_j} = \frac{N_j}{T\varepsilon_j} = \frac{N - N_{bkgj}}{T\varepsilon_j} \qquad (3)$$

were c_j is the contribution of the *j*-th isotope to the integrated spectrum counts; ε_j is the counting efficiency for the *j*-th isotope at a quenching parameter of the sample; N_{bkg} is the background counts integrated over all channels; and $N_{bkgj} = N - N_j$ is the total integral account of all radionuclides in the sample, including the blank sample background, after subtraction of the *j*-th radionuclide.

The combined standard uncertainty of *j*-th isotope activity (for $T_{sample} = T_{bkg}$) is

$$\frac{\sigma_{A_j}}{A_j} = \sqrt{\frac{N + (N - N_j)}{(N - (N - N_j))^2} + \frac{\sigma_{\varepsilon_j}^2}{\varepsilon_j^2} + \frac{\sigma_{c_j}^2}{c_j^2}} = \sqrt{\frac{2N - N_j}{N_j^2} + \frac{\sigma_{\varepsilon_j}^2}{\varepsilon_j^2} + \frac{\sigma_{c_j}^2}{c_j^2}} \tag{4}$$

where $N_j = (N - N_j) \times c_j$ is the contribution of the determined nuclide to the integral count of the sample, σ_{c_j} is the uncertainty resulting from the fitting procedure, and σ_{ε_j} is the uncertainty resulting from the counting efficiency determination (combined, includes statistical, standard solution certification, standard solution mass, and quenching parameter determination uncertainty).

Method Realization

The algorithms described above for splitting a LS spectrum into its components (radionuclide identification), and also to calculate their activities, uncertainties, and the lower limit of detection (LLD) in the optimized region of spectrum, are realized using the RadSpectraDec software. The program is written in C++ and operates with Windows® 98/ME/NT/2000/XP/2003. This program is intended for mathematical processing of spectra measured using LS counters, in particular Tri-Carb (Packard), Quantulus (Malinovsky et al. 2002b), Guardian (Wallac), and Triathler (Hidex). The program can be also modified and used for other types of counters.

RESULTS AND DISCUSSION

LS spectrometry for rapid analysis of radionuclide composition (screening) of liquid samples is most useful for the following: control of radionuclide composition and contents in technological media, monitoring liquid discharges for manufacturers that produce radionuclides of natural and manmade origin, and radiation monitoring of the potable water supply. Below, the results of rapid radionuclide examinations are demonstrated with examples of various liquid samples.

Liquid Radioactive Wastes

Results of LS spectrometric analysis of the radionuclide composition of liquid radioactive wastes (LRW) with the application of extracting-scintillation techniques are given in Figure 2 and Table 1. Due to the high content of beta emitters in the sample, it was not possible to discriminate alpha emitters directly by processing the spectrum of the initial sample. To assess the concentration of alpha-emitting radionuclides represented by polyvalent elements, the radionuclides were separated and concentrated by extraction with a HDEHP-based scintillator. As a result of the screening, it became clear that [93]Mo is one of the basic radionuclide components of this waste. Waste identification as an alternative to LSC methods is practically impossible (E_{photon} = 16–18 keV).

Results of the examination of another LRW radionuclide composition in certain treatment stages of LS spectrometry, gamma spectrometry, and by radiochemical methods are shown in Figure 3 and Table 2.
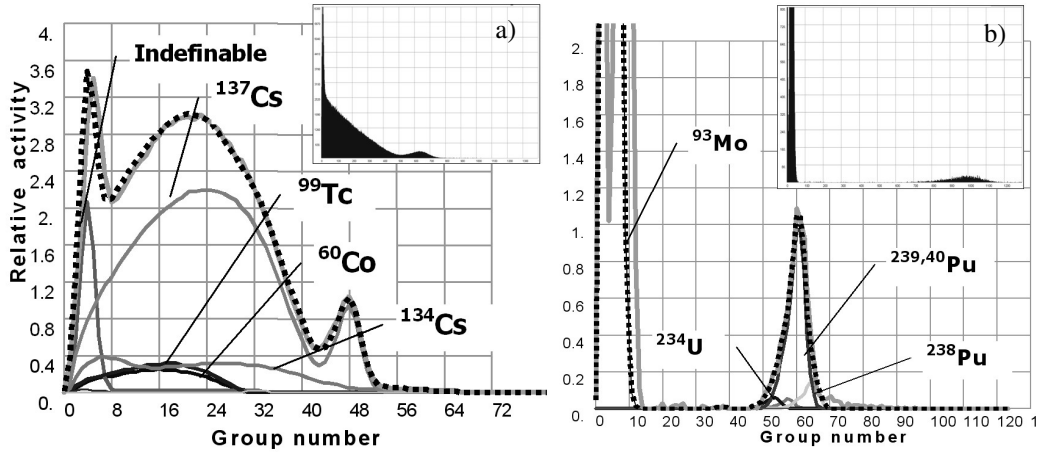
Figure 2 LS spectra decoding for liquid radioactive waste (LRW) samples from an industrial reactor unit: a) initial composition; b) after extraction with HDEHP.

Table 1 Basic radionuclide composition of liquid radioactive waste (LWR) from an industrial reactor unit.

	LRW, initial composition	LRW, alpha emitters
Sample preparation	1 mL + Ultima Gold AB	5 mL 0.1M HNO_3 solution, extraction with 0.2M HDEHP in Ultima Gold F
Radionuclide[a]	^{60}Co: 610 ± 40 Indefinable: 1000 ± 200[b] ^{99}Tc: 620 ± 50 ^{134}Cs: 1330 ± 40 ^{137}Cs: 5810 ± 60	^{93}Mo: 1100 ± 50 $^{239+240}$Pu: 50 ± 4 ^{238}Pu+^{241}Am: 5 ± 2

[a]Specific activity (Bq/L), ±2 σ.
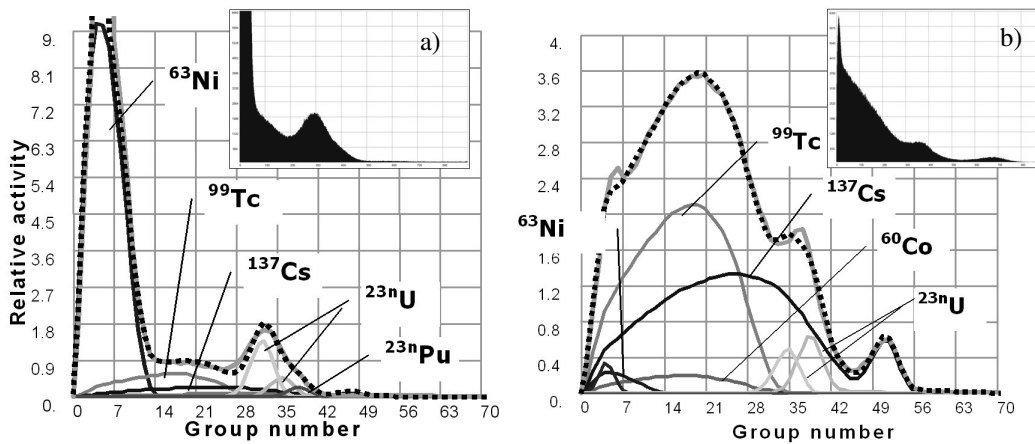[b]Unknown efficiency.

Figure 3 LS spectra decoding for liquid radioactive waste (LRW) samples at certain stages of treatment: a) initial composition; b) after 2nd stage of purification.

Table 2 Determination of radionuclide composition of liquid radioactive waste (LRW) at certain stages of treatment.

Analytical technique	LRW, initial composition		LRW, after 2nd stage of purification	
	Screening[a]	Radiochemistry + gamma spect.	Screening[a]	Radiochemistry + gamma spect.
Analyzed volume (mL)	1	50	2	50
Radionuclide	Specific activity (Bq/L), ±2 σ			
^{63}Ni	63,500 ± 2000	60,000 ± 1500	700 ± 200	1100 ± 100
^{99}Tc	2500 ± 200	1200 ± 100	10,500 ± 700	9200 ± 500
^{137}Cs	1100 ± 100	2850 ± 250	9400 ± 400	11,000 ± 700
^{60}Co	8900 ± 300	11,400 ± 500	450 ± 100	200 ± 20
^{90}Sr	100 ± 40	110 ± 10	300 ± 50	400 ± 40
ΣU	7200 ± 200	12,700 ± 700	1500 ± 200	1200 ± 100
ΣPu	1000 ± 100	4650 ± 250	≤30	≤10

[a]Results of the γ spectrometry are taken into account during LS spectra processing.

Taking into account the submitted examples, it is clear that this approach in combination with gamma spectrometry allows one to rapidly define principal components even in sufficiently complicated radionuclide compositions of technological samples.

Water from Control Drills, Liquid Leakages

Examples of application of LS screening for monitoring of liquid discharges are given in Figure 4 and Table 3. Calculated contents of main radionuclide components in the counting source were
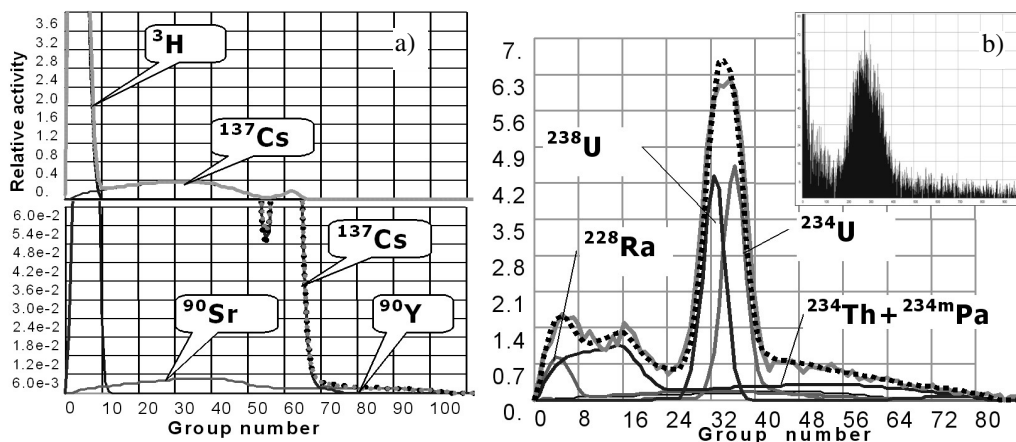

Figure 4 LS spectra decoding for the liquid samples taken from monitoring boreholes: a) in the region of subterranean nuclear tests; b) a region with increased uranium content in rocks.

Table 3 Determination of radionuclide composition of water from monitoring boreholes.

Sample code	Analyzed volume (mL)	Specific activity (Bq/L), ±2 σ			
		^{234}U	^{238}U	^{226}Ra	^{228}Ra
St 2, screening	7	12 ± 2	14 ± 2	1	5.2 ± 1.1
St 2, radiochem.	500	11.8 ± 1.3	10.6 ± 1.2	0.96 ± 0.05	0.46 ± 0.03

obtained in a 1-mL borehole water sample: $^3H = 5700 \pm 60$ Bq; $^{137}Cs = 450 \pm 5$ Bq; $(^{90}Sr+^{90}Y) = 9 \pm 2$ Bq. Liquids in which the content of radionuclides exceeds a PL^{water} (permissible levels of radionuclides in potable water) on the order of 1 magnitude or more may be qualified as radioactive wastes (Ministry of Health Protection 1999). A sample volume of 1–20 mL is enough to ensure the required sensitivity for analysis and thus to come to reliable conclusions on this subject.

Potable Water

During radiation monitoring of the potable water supply, the control levels of specific volumetric activity concentrations are determined as 0.1 Bq/L for alpha emitters and 1 Bq/L for beta emitters. When those values are exceeded, it is necessary, according to the methodical instructions, to sequentially determine the number of radionuclides via radiochemical methods in order to compare their specific activities with the Russian limits (Ministry of Health Protection 1999) (Table 4).

Table 4 Permissible levels of radionuclides in potable water
(Ministry of Health Protection 1999: Supplement-2, NRS-99).

Radionuclide	Half-life	PL^{water} (Bq/kg)
^{90}Sr	29.1 yr	5.0
^{210}Pb	22.3 yr	0.2
^{210}Po	138 d	0.12
^{224}Ra	3.66 d	2.1
^{226}Ra	1600 yr	0.5
^{228}Ra	5.75 yr	0.2
^{228}Th	1.91 yr	1.9
^{230}Th	7.70×10^4 yr	0.66
^{234}U	2.44×10^5 yr	2.9
^{238}U	4.47×10^9 yr	3.1

Examples are given below of LS analysis on natural waters with increased contents of alpha emitters (Figure 5 and 6). Results of LS spectrometry and comparative radiochemical analyses are listed in
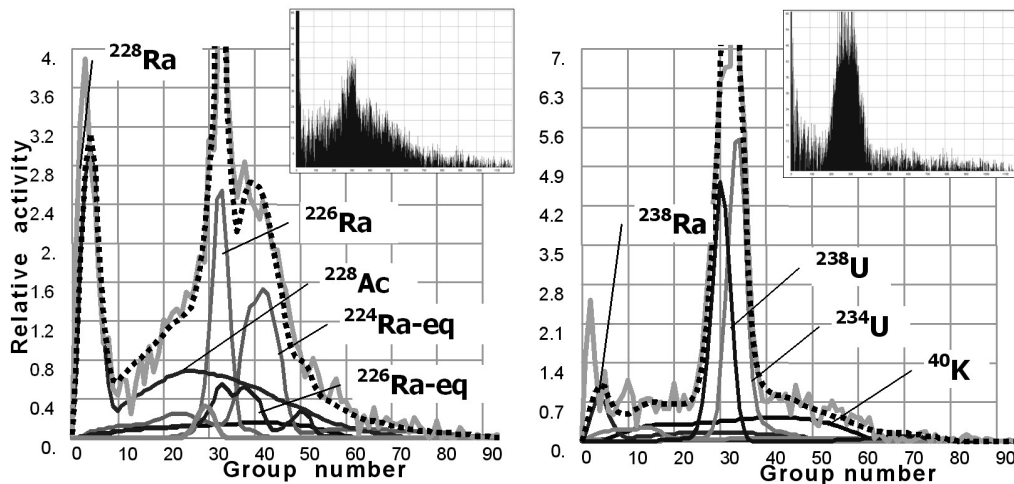

Figure 5 LS spectra (after background correction) of potable water. Left side: Lipetsk district, sample S 1; Right side: Moscow district, sample V 6. Initial (inset) and decoded (lower) spectra using the RadSpectraDec program.
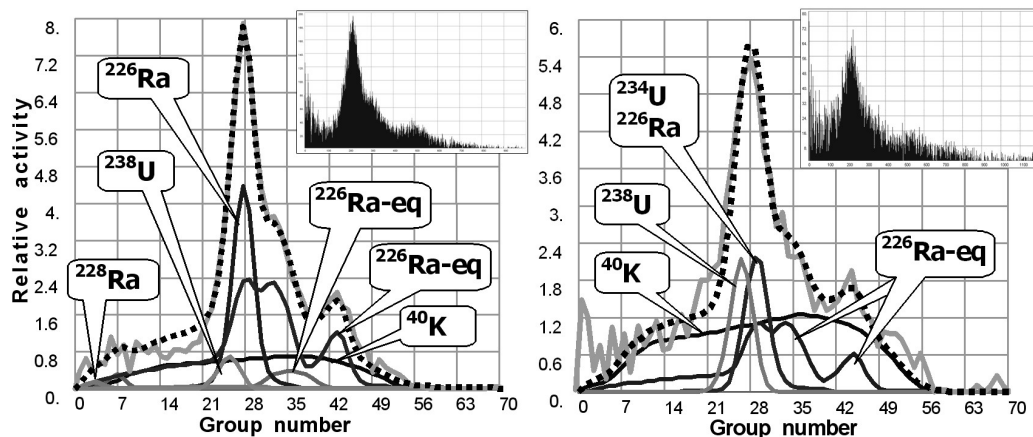
Figure 6 LS spectra (after background correction) of potable water. Left side: Tver district, sample ZV 2; Right side: Vladimir district, sample RN 1. Initial (inset) and decoded (lower) spectra using the RadSpectraDec program.

Tables 5 and 6. Sample preparation included evaporation of the initial sample in volumes of 300 mL up to 5 L.

Table 5 Determination of radionuclide composition in potable water samples.

Sample code	Analyzed volume (mL)	Specific activity, ±2 σ (Bq/L)				
		^{234}U	^{238}U	^{226}Ra	^{228}Ra	^{210}Po
S 1, screening	300	≤0.05	≤0.05	0.19 ± 0.05	0.32 ± 0.04	0.04 ± 0.02
S 1, radiochem.	1500	0.03 ± 0.01	0.03 ± 0.01	0.18 ± 0.05	0.33 ± 0.02	0.015 ± 0.005
V 6, screening	300	0.37 ± 0.04	0.30 ± 0.03	≤0.02	0.07 ± 0.03	≤0.02
V 6, radiochem.	1000	0.363 ± 0.036	0.245 ± 0.025	0.019 ± 0.009	0.004 ± 0.002	≤0.01

Table 6 Determination of radionuclide composition in potable water samples.

Sample code	Analyzed volume (mL)	Specific activity, ±2 σ (Bq/L)				
		^{234}U	^{238}U	^{226}Ra	^{228}Ra	^{210}Po
ZV 2, screening	300	—[a]	0.06 ± 0.03	0.54 ± 0.06	0.04 ± 0.02	0.03
ZV 2, radiochem.	1500	0.036 ± 0.010	0.018 ± 0.005	0.65 ± 0.08	0.015 ± 0.008	0.01
RN 1, screening	300	0.06 ± 0.03[a]	0.06 ± 0.03	0.09 ± 0.03	0.02	0.02
RN 1, radiochem.	1000	0.05	0.05	0.18 ± 0.04	0.01	0.01

[a]Not separated from ^{226}Ra during spectra processing.

From the data, one can see that in both samples the control level of specific activity of alpha emitters (0.1 Bq/L) is exceeded considerably. According to the results from screening sample S 1, the alpha activity was almost completely defined by ^{226}Ra, ^{224}Ra, and their progeny, while in sample V 6 the alpha activity is defined by the U isotopes. An exceeding PL^{water} value for ^{228}Ra in the first sample was confirmed by radiochemical analysis. Determination of uranium by the isotopic dilution method using high-resolution alpha spectrometry shows values of activity concentrations similar to those of the screening method. In both samples, an absence of noticeable concentrations of ^{210}Po (the most toxic radionuclide for potable waters) was confirmed by radiochemical separations.

LS screening has shown that an excess in both samples in the control level of alpha emitters was caused mainly by the presence of ^{226}Ra and its progeny. An exceeding PL^{water} value for ^{226}Ra in sample ZV 2 was confirmed by radiochemical analyses. It is necessary to note that in the listed examples, preliminary screening investigations of potable water via LS spectrometry have avoided several time-consuming radiochemical procedures, which in traditional examinations should be used randomly, taking into account all possible components.

CONCLUSION

We present a method for simultaneously identifying and quantifying the radionuclide composition of complex radionuclide mixtures by using LSC. This method is based on the modeling of initial spectra transformed by a convolution into group form by the superposition of individual reference spectra taken from an existing nuclide library. This process is realized in different versions of the expert software package RadSpectraDec.

During long-term, intensive use of the described method, rapid LS analyses were applied for environmental and technological samples of various origin. In a number of cases, our method allows us to avoid several time-consuming radiochemical procedures, thus allowing one to quickly assess the composition of basic alpha- and beta-emitting radionuclides, or at least to determine whether they exceed control levels for particular radionuclides, a primary aim of radiation monitoring. In complicated cases, it is possible to make decisions about using additional examinations based on high-resolution alpha spectrometry or beta spectrometry with preliminary chemical separations. At the same time, the activity of isotopic tracers is also assessed. The precision and reliability of the described method depends on the quality of the reference solutions, the chemical and radionuclide mixture composition, and the counting statistics.

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