# Development and Application of Collimated Spectrometric Systems for the Characterization of Radioactive Contamination of Decommissioned Facilities – 15030

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# ABSTRACT

Different approaches use collimated spectrometric systems for the characterization of radioactive contamination of various objects: soil, constructions decommissioned facilities. From time of works on elimination of Chernobyl accident and other large scale contamination accidents due radioactive dawnfalls we start to use "CORAD" method which gives values of surface contamination of soil (in Ci/m<sup>2</sup>) and depth of soil contamination (in MFP or cm) using measurements with collimated spectrometric detector NaI(Tl). The realizations of method for measuring of surface contamination of walls, floors using spectrometric detectors of various types were carried out in last years. We present the main results of the work - the upgraded detector for measuring contamination in wells and concrete measurements in contaminated areas of engineering rooms. Realization of method using different types of spectrometric detectors scintillating NaI(Tl) and LaBr3, semiconductor - HPGe and room temperature CdZnTe. Different types of detector are the most suitable for different conditions of measurements. The results of measurements of contaminated soil, sediment, concrete construction floor and walls in the rooms are presented.

### **INTRODUCTION**

The measurements of surface soil contamination due to radiation dawn-falls have long history and is relatively labour-consuming task [1,2]. In measurements of Chernobyl accident and other large scale contamination due dawn-falls we start to use "CORAD" method which gives values of surface contamination of soil (in Ci/m2) and depth of soil contamination (in MFP or cm) using measurements with collimated spectrometric detector NaI(Tl) [3-6].

The practice of measurements in different conditions: Given the heterogeneity of distribution of contamination, additional background from other contaminated objects in sites. We consider the possibility of extending of carrying out measurements using a collimated detectors for scanning systems. Possibility of modifying the method for estimating the distribution of species contamination in depth - exponential, linear profiles. Conducted research on methods of localization of deep contamination of concrete structures and pipelines them in search of non-destructive methods and remote diagnostics. We investigated the possibility of using a combination of measurements with collimated detector and borehole collimated detectors for measurements on localization pipelines.

Studied non-destructive methods were used for work in the contaminated areas of engineering premises of NRC Kurchatov Institute. The measurement results are very efficiently processed using the geostatistical methods, which allow us to give the best spatial interpolation and statistical evaluation of measurement data. The data presented are treated using as Open Source the statistical package R and both with the help of a commercial product Kartotrak (Geovarians, France).

# **MEASUREMENTS, INSTRUMENTS AND METHODS**

For the measurement of radionuclide contamination of the concrete floor was selected one room of the process facilities at NRC "Kurchatov Institute". In this room a long time is the setting in which work was conducted with radioactive liquids. As a result, the floor has a significant spill contamination. Also located in the floor pipe line, which is also radioactive liquid siphoned. The exact location of pipelines is unknown. To test whether the search contaminated industrial pipelines occurring in the concrete at

shallow depth (15-20 cm), a detailed examination of the technological facilities in the preparatory work for the room to rehabilitation.

For measurements of radionuclide contamination of the concrete used collimated spectrometric detector with a scintillator LaBr3. The spectrum was recorded with multichannel analyzer InSpector 1000. At each point of measurement, the detector mounted on a stand, measured two spectra with an open and with a closed collimator. Processing of the spectra was carried out with a special program based on the method of "CORAD" [7]. A variant of this technique for the detector with a scintillator LaBr3 - «Korad-LaBr3» was developed previously [8]. This algorithm gives consistent results when multiple gamma emitting nuclides are present.



Fig. 1 The spectrometric system «Corad-LaBr3» in the process of calibration measurements. 1 - spectrum analyzer "Inspector 1000", 2 - scintillating LaBr3 detector with a lead collimator and shielding, 3 - collimator shutter, 4 - flat gamma-ray source with known surface activity.

The first step of the measurement procedure was to calibrate the instrument on the plane (film) sources with known surface activity. Used for this purpose film sources of <sup>137</sup>Cs and <sup>60</sup>Co with surface activities  $A_0$ = 20.5 µCi/m<sup>2</sup> (759 kBq/m<sup>2</sup>) and A0  $A_0$ = 12.1 µCi/m<sup>2</sup> (448 kBq/m<sup>2</sup>), respectively. The procedure of the spectrometer calibration is shown in Fig. 1. The task of calibration is to determine the coefficients  $\alpha$ ,  $\beta$  and  $\alpha'$ ,  $\beta'$  which correlate integrals in spectral range with spatial activity and material attenuation of radiation of corresponding nuclide.



Fig. 2 Spectra obtained in measurements on the fine mesh 0.4 x 0.4 m. Difference spectrum is used for evaluation of  $^{137}$ Cs and  $^{60}$ Co contamination.

### Result of survey of concrete floor

After the calibration of the spectrometric system "Corad-LaBr3" radiometric survey was carried out a concrete floor in a room with contaminated reactor facility. Based on the results of these measurements are carried out measurements of spectra processing program that implements the method described above evaluate the performance of contamination of <sup>137</sup>Cs and <sup>60</sup>Co. On the floor of this room was chosen rectangular area measuring  $3.6 \times 2.4 \text{ m}$ . In this are were two radiometric survey - one on the grid with a coarse step of 120 cm, and the with fine step of 40 cm

As mentioned above, at each point two measurements - with and without shutter were carried out. Fig. 2 shows the result of the measurement at point #7 fine grid.

The figure 2 are clearly opposed the extent and nature of concrete contamination by <sup>137</sup>Cs and <sup>60</sup>Co. There are characteristic differences in magnitude pulses detected, and the nature of the difference spectra. The exposure time for each measurement point with and without shutter, were identical and equal to 5 minutes. Details of contamination for <sup>60</sup>Co and <sup>137</sup>Cs obtained from measurements on coarse and fine grids are given in Tables 1 and 2.

Table 1. Characteristics of concrete contamination by <sup>1</sup>	<sup>137</sup> Cs and <sup>60</sup> Co obtained from measurements on a
large grid 120 x 120 cm	

	A <sub>Cs</sub> ,	Z <sub>Cs</sub> ,	$L_0$ ,	$L_1$ ,	A <sub>Co</sub> ,	Z <sub>Co</sub> ,	$L_{0Co}$ ,	$L_{1\mathrm{Co}}$ ,
	$\mu Ci/m^2$	mfp	mfp	mfp	$\mu Ci/m^2$	mfp	mfp	mfp
1.	12600.0	3.21	0.71	2.5	2810.0	5.85	3.35	2.5
2.	735.0	0.72	0.0	0.72	300.0	5.85	3.35	2.5
3.	242.0	0.51	0.0	0.51	156.0	4.09	1.59	2.5
4.	177.0	0.35	0.0	0.35	349.0	4.75	2.25	2.5
5.	1570.0	4.22	1.72	2.5	179.0	5.85	3.35	2.5
6.	393.0	1.42	0.0	1.42	98.1	1.82	0.0	1.82
7.	481.0	1.3	0.0	1.3	82.5	1.81	0.0	1.81
8.	246.0	1.22	0.0	1.22	307.0	3.74	1.24	2.5
9.	41.7	0.66	0.0	0.66	3.56	0.0	0.0	0.0
10.	224.0	0.83	0.0	0.83	9.44	0.58	0.0	0.58
11.	431.0	0.33	0.0	0.33	34.2	0.2	0.0	0.2
12.	2600.0	1.17	0.0	1.17	924.0	3.08	0.58	2.5

Table 2. Characteristics of concrete contamination by  $^{137}$ Cs and  $^{60}$ Co obtained from measurements on a large grid 40 x 40 cm

							r	
	$A_{Cs}$ ,	$Z_{\rm Cs}$ ,	$L_0$ ,	$L_1$ ,	$A_{Co}$ ,	$Z_{\rm Co}$ ,	$L_{0Co}$ ,	$L_{1Co}$ ,
	$\mu Ci/m^2$	mfp	mfp	mfp	$\mu Ci/m^2$	mfp	mfp	mfp
1.	7410.0	5.56	3.06	2.5	0.0	0.0	0.0	0.0
2.	268.0	0.04	0.0	0.04	0.0	0.0	0.0	0.0
3.	74.9	0.26	0.0	0.26	148.0	5.85	3.35	2.5
4.	445.0	0.7	0.0	0.7	131.0	5.02	2.52	2.5
5.	61900.0	3.6	1.1	2.5	21500.0	5.85	3.35	2.5
6.	81.4	0.1	0.0	0.1	524.0	5.85	3.35	2.5
7.	61.1	0.0	0.0	0.0	439.0	5.85	3.35	2.5
8.	555.0	1.02	0.0	1.02	363.0	5.46	2.96	2.5
9.	19.0	5.19	2.69	2.5	799.0	5.85	3.35	2.5
10.	120.0	0.07	0.0	0.07	7.08	0.0	0.0	0.0

11.	106.0	0.68	0.0	0.68	24.3	0.88	0.0	0.88
12.	736.0	0.43	0.0	0.43	77.9	1.08	0.0	1.08
13.	52.3	1.86	0.0	1.86	0.0	0.0	0.0	0.0
14.	25.7	0.0	0.0	0.0	6.81	2.1	0.0	2.1
15.	22.0	0.0	0.0	0.0	24.9	3.62	1.12	2.5
16.	368.0	0.89	0.0	0.89	70.4	0.13	0.0	0.13

Surface distributions of <sup>137</sup>Cs and <sup>60</sup>Co activity, the depth of penetration into the concrete Z, obtained from the results of radiometric surveys for large and small grid are shown in Fig. 3 and 4.



Fig. 3. The surface activity (A) and penetration (B) of radionuclides  $^{137}$ Cs in concrete measured on a fine grid 0.4 x 0.4 m (16 points)



Fig. 4. The surface activity (A) and penetration (B) of radionuclides  $^{60}$ Co in concrete measured on a fine grid 0.4 x 0.4 m (16 points)

### Measurements for localization of deep contamination of concrete structures and pipelines

Consideration of the approach to search deep contamination carried by the example of the two main nuclides present in the contaminated concrete of objects decommissioned - <sup>137</sup>Cs and <sup>60</sup>Co. The developed algorithm allows the consideration of other nuclides with sufficient energy of gamma lines (the algorithm runs up to the penetration depth of 3-4 mean free path of the corresponding gamma rays). Also, when you transfer the algorithm to other nuclides to be considered and some features of the problem - the ratio of the activities, the energy location of the used lines of gamma-radiation.



Fig. 5 Exterior of the floor in the rooms where the measurements of radioactive contamination of the concrete were carried out.

The floor of room where the measurements have been was marked by a square grid with a pitch of 0.5 m. The result was 175 points for measurements. Another three points were selected in the next room, where the concrete floor was opened, removed part of the pipeline and were capped with clean gravel. Remains below the level of contamination of the concrete pipe is accurate rapper when the depth of contamination of the concrete, the corresponding location of the pipeline.

### **Results of measurements**

The obtained primary data measurements - two spectrum at each point. After mathematical processing of each pair of spectral data are obtained on the characteristics of contamination. In our case, we used the model the presence of two nuclides - <sup>137</sup>Cs and <sup>60</sup>Co. The processing of the spectra was obtained 8 values characterizing contamination of the concrete at each point. This surface activity of <sup>137</sup>Cs in pCi/m2 (the variable CsA) penetration depth of <sup>137</sup>Cs contamination (CsZ), the thickness of the layer of pure <sup>137</sup>Cs (CsL0), the thickness of the contaminated layer for <sup>137</sup>Cs (CsL1) and similar values for radionuclide <sup>60</sup>Co - CoA, CoZ, CoL0, CoL1. Dimensions - depth and thickness of the layers are measured in mean free paths (MFP) of gamma rays with energy of 662 keV. For concrete with medium density, this value is about 7 cm.

The data in Figure 15 for the surface activity of <sup>137</sup>Cs and <sup>60</sup>Co are shown in the map of the room. It can be seen that at one point (X in Fig. 14) the data are nearly 100 times different from those at other measurement points. This point is located adjacent the opening leading, presumably, a buried tank. According to the measurements assessed total value of activity in the contained concrete floor of the room. The total activity of <sup>137</sup>Cs is 1.56 GBq, and the total activity of <sup>60</sup>Co is 0.33 GBq, the volume of contaminated concrete is 6.75 m3. A large part of this volume is contaminated by <sup>60</sup>Co.



CoA

CsA



Fig. 6 data on the distribution of surface activity  $^{60}$ Co (top) and  $^{137}$ Cs (bottom). Activity is proportional to the size of the circle. The scale is logarithmic.

# **Processing of measurement results**

Figures 7 and 8 show pure thicknesses of layers of contaminated concrete for <sup>60</sup>Co and <sup>137</sup>Cs obtained after processing the spectra. The layer thickness are in units MFP (mean free pass) of the gamma-quantum with 662 keV in the concrete.



CoL1



Fig. 7 The data of measuring the thickness of the pure (top) and contaminated (bottom) layer of concrete for  ${}^{60}$ Co. The layer thickness in units of MFP.



CsL1



Fig. 8. The data of measuring the thickness of the pure (above) and contaminated (bottom) layer of concrete for <sup>137</sup>Cs. The layer thickness in units of MFP.

Both the maps of thickness distribution of contaminated layers and statistical analysis show the average value of the penetration depth for <sup>60</sup>Co is more than for <sup>137</sup>Cs. Furthermore, the areas in which there is a deep penetration into the concrete of radionuclides <sup>60</sup>Co and <sup>137</sup>Cs not coincide. Perhaps this is due to the ambiguity in determining the thickness of the contaminated layer (and the total depth of penetration) in some cases.

#### Analysis and interpretation of the results

Ambiguity in determining the thickness of the contaminated layer (and the total depth of penetration) in condition when there is a conditionally clean (low-contaminated) layer of material creates a certain ambiguity in the search of buried contaminants. In this case, the depth of the contamination is determined with the ambiguity the model used in the calculations by the "CORAD" method.



Fig. 9 The dependence of ratio of the integrals C / Ph on penetration depth of contamination Z for the different types of the contamination depth distribution.

The graphs from Fig. 9 explained what is connected with the ambiguity of the depth estimation. If the measured ratio of the integrals obtained count rates in the Compton area and photopeak C / Ph more critical value shown in red dotted line, then there is a "quasi" clean layer above contaminated layer. Thus by the ratio C/Ph is largely affected by the thickness of the clear layer, and selecting the thickness of the contaminated layers gives the uncertainty.

To reduce the effect of this uncertainty in the identification of the location and depth of contamination pipelines we introduce a new feature of the penetration depth of contamination, combining data for cobalt and cesium - generalized penetration of contamination:

$$Z^* = (1/(1+k)) (CsL0 + 0.5 CsL1 + k(CoL0 + 0.5CoL1))$$

Maps of the generalized penetration of contamination for two values of the parameter k are shown in Figure 10.



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Fig. 10 The maps of the generalized penetration of contamination -  $Z^*$  constructed for k = 1 (top) and k = 2 (bottom)

As can be seen from the maps in Figure 10, the introduction of the value of the generalized penetration of contamination enables to make an assumption about the location of pipelines. Rather, they are located where held vertical and horizontal lines on the map, along which the random nature of the penetration depth of contamination in the concrete turns into a regular linear structure.

The data obtained present the three-dimensional spatial distribution (along the surface of the premises and depth) of the two values of the specific activity - nuclide Cs-137 and Co-60. Depth distribution is not smooth and piecewise character - the thickness of the contaminated layer and the thickness of the concrete

zz11

value-standing (if any) over the contaminated layer. Joint submission of data will help to identify spatial patterns in the distribution and give suggestion on the location of pipelines in the area. For a threedimensional representation of data a program of spatial visualization of complex data. The program provides an opportunity to represent piecewise distribution of several variables defined on a twodimensional grid of arbitrary form. The program is written in VRML, which creates the necessary threedimensional scene. The display computer displays arbitrary scene viewer VRML files. We used a Cortona3D Viewer. Spatial presentation of measurement data contamination of the concrete floor in the room is shown in Figure 11.



Fig. 11. Spatial presentation of measurement data of contamination of the concrete floor in the room. The color palette shows the levels of specific activity of Cs-137 and Co-60 between the minimum and maximum values.

The resulting three-dimensional visualization of data allows the interactive analysis. According to the results, you can make an idea about the possible spatial arrangement of process piping in the surveyed area. Figure 12 shows the results of online search possible location of pipelines in the area on the spatial representation of measurements of contamination of the concrete floor. Set view of the distribution of contamination from the "inside" of the concrete floor, which is well traced line piping arrangement.

### DISCUSSION

The results of studies of measurements of concrete contamination and the localization of deep contamination of concrete structures and pipelines based on remote and non-destructive methods are presented..



Fig. 12 Interactive search of pipelines location in the area using the 3-D representation of measurements of contamination of the concrete floor. The view of the distribution of the contamination is shown from the "inside" of the concrete floor. The grid lines are removed for clarity. Red lines - the proposed location of pipelines.

To test whether the search contaminated industrial pipelines occurring in the concrete at shallow depth (15-20 cm), a detailed examination of the technological premises in heterogeneously contaminated concrete floor in the preparatory work for the room to rehabilitation. The measurements were carried out on an area of about 50 m<sup>2</sup> using a collimated spectrometer based detector LaBr3, the results were processed by the method of "Korad." Measurements were carried out at 178 points. The results of measurements were defined penetration depth nuclides <sup>137</sup>Cs and <sup>60</sup>Co in concrete (total depth, thickness the contaminated layer, relatively clean upper layer) and the corresponding surface activity. Upon receiving the initial data determine the specific activity of contamination of the total activity of contamination and the expected volume of waste during the rehabilitation work in the room. Spend the development of a program of spatial visualization of the integrated presentation of all data obtained after processing the measurement results. The resulting visualization allows interactive analysis. According to the results, you can get a perspective view of a possible arrangement of process piping in the surveyed area. A computer program controls the spectrometer system and measurement data processing.

To increase productivity of mapping it useful to produce automatically scanning system as Gammalocator system [9]. The method of determination of the thickness of the pure and contaminated layer of is reliable when measurements are carried out in one standard position, say perpendicular to the surface. So this system must produce mechanical moving of measurement part – collimated detector.

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