Characterisation of rooms and buildings concrete surfaces for decontamination and free-release at NPP A-1, Slovakia

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Abstract:
Dominant contamination at NPP A-1 is caused by long-living ^{137}Cs that may occur in concrete structures. Therefore, characterisation is required for determination the method and range of decontamination (before decontamination) as well as for scope and range of free release measurement (after decontamination).
The base of characterisation is monitoring of total activity in the structure per unit surface area [Bq/cm^2]. This approach is in compliance with international recommendation RP-113. Measuring equipment including hand-held gamma spectrometry assembly with scintillation LaBr 1.5” x 1.5” detector was applied. The measuring assembly was certified by Slovak Metrology Institute as measuring instrument of 15% accuracy class.
The proposed method, estimation of measurement uncertainty and other support characterisation methods as well as results and experience from a pilot in situ monitoring of a large concrete tank are discussed in more detail in this paper.

Keywords: contamination depth, density of monitoring network, in situ monitoring, total activity in the structure per unit surface area

Introduction

Characterisation of concrete surfaces in Slovakia is focused mainly to NPP A-1 that is under 2nd stage of decommissioning project. The NPP A-1 has been shut downed non-standard after nuclear accident with fuel core melting. Consequently, dominant contamination is caused by long-living ^{137}Cs that may occur in concrete structures. The aim of building structures decommissioning is their decontamination and consecutive demonstration of fulfilment the requirements for free release in compliance with national legislative, before demolition of these building structures. Therefore, characterisation is required for determination of methods and range of decontamination (before decontamination) as well as for scope and range of free release measurement (after decontamination).
Characterisation measurements shall be performed in compliance with national legislation by means metrologicaly certified measuring instruments.

Legislative requirements for free release into the environment

The free release into the environment in Slovak republic is regulated by Regulation of the government of SR No 345/2006. From point of view of this act the contamination of the concrete structures is considered to be volumetric contaminated material. These free released materials have to fulfil prescribed clearance limits for massic activity of important radionuclides as well as the sum rule:

$$\sum_{i=1}^{n} \frac{A_i}{A_{Li}} \leq 1$$

where
A_i is mass activity of *i*-th radionuclide and
A_{Li} is clearance limit for mass activity of *i*-th radionuclide.

For example, clearance limit of dominant ^{137}Cs is 300 Bq/kg.

Considering that the contamination mostly occurs in the slim surface concrete layer (thickness of 0 to 20 mm) from the point of view the time and cost consumption it is more advantageous to remove the contaminated surface layer and consequently to free release the concrete structure as denuclearised building.
In our case an approach was chosen that is in compliance with buildings release concept according to the international recommendation RP-113. This approach is based on the determination of the total activity in the structure per unit surface area (TAUSA). TAUSA have to be determined in the full depth profile of contaminated concrete layer. Clearance limit of TAUSA for single ^{137}Cs is 10 Bq/cm^2. This value has to be corrected taking to account other important radionuclides occurring in concrete structure.
Fulfilling the requirements for TAUSA in compliance with RP-113 is leading to fulfil the national legislative requirements for massic activity.

Characterisation measurements

Characterisation process of concrete surface includes the combination of various *in situ* non destructive monitoring methods as well as the destructive sampling methods followed by a laboratory analysis.
The base of characterisation is monitoring of total activity in the structure per unit surface area [Bq/cm^2]. This measurement based on *in situ* scintillation gamma spectrometry is in more detail described farther in separate chapter.
Particular characterisation methods are described below.

Preliminary dose rate survey

The first step of characterisation is preliminary dose rate survey. This monitoring is necessary for identification and removing of a "hot spots". Operational dose rate threshold for "hot spot" identification on monitored concrete surface was proposed on level 2 µSv/h. That means the decrease of an external interfering radiation and workers dose burden.

Determination of effective contamination depth

Determination of effective contamination depth is required for choose of adequate decontamination method. The effective contamination depth is determined by sampling (core or common drill) and/or by *in situ* semiconductor gamma spectrometry.

Sampling by core and common drill

Taken samples of core drills are segmented by means of a circular saw and particular segments are analyzed by laboratory semiconductor gamma spectrometry. The thickness of the particular segments is limited to minimum 5 mm. The homogenous activity distribution in the particular segment samples is checked by measurement of the both segment sides. If the difference of the response from first and second side exceeds 20 % the activity distribution is considered to be inhomogenous. In this case the segment sample is powderized by the hydraulic press equipment. The obtained dust sample is homogenised and subsequently measured. The depth in which the measured specific activity does not exceed the free release limits required by legislative (e.g. 300 Bq/kg for ^{137}Cs) is considered as effective depth of contamination.
For increase of representativeness two or three parallel core samples are taken from the same site.
Analysing of the core drill samples is important to demonstrate that not contamination occurs inside the concrete structures. The sampling by core drill across the entire width of the wall is applicable in the case when a second side of the wall is not available for sampling or *in situ* measurement.

The sampling by using common drill requires the set of drills with various diameters. Firstly the drill with maximum diameter is used. Next bore on the same site is carry out by drill with smaller diameter in comparison with the previous one. In this manner obtained concrete dust samples are less difficult to laboratory processing and analysing but acquired quantity of sample is smaller in comparison with core drill.
The acquired concrete samples are used to determination of specific activity of the hard to detectable radionuclides (HDRN) like ^{90}Sr, ^{241}Am or isotopes of Pu that are alpha or beta emitters only.

Non destructive *in situ* method for contamination depth estimation

For the effective contamination depth estimation and activity determination in concrete structure of surface the non destructive measuring methods may be applied. Method is based on the *in situ* semiconductor gamma spectrometry with shielded HPGe detector. Estimation of the effective contamination depth is based on various linear attenuation coefficients for the various energies of radiation in the concrete structure. Effective contamination depth is determined by the ratio of net full energy peak area of 661.6 keV (^{137}Cs) gamma ray and net full energy peak area of 32 keV X-ray (^{137}Cs). Detection efficiency of 661.6 keV and 32 keV of ^{137}Cs depends on the contamination depth and for various depths can be determined empirically or by means of Monte Carlo simulations. Disadvantage of this method is difficult manipulation and transportation of the spectrometry assembly in not available sites of controlled area (heavy shielding, fill up the liquid nitrogen for detector cooling, etc.).

Monitoring of surface contamination

For the surface contamination monitoring of a concrete structures a hand-held measuring instruments based on the sum beta radiation detection are used. These instruments are commonly equipped with gas proportional or plastic scintillation detectors with sensitive area less 200 cm^2. Due to the beta radiation detection they are able to measure contamination of thin surface layer (about 1 mm), only. The thickness of the effectively monitored surface layer is corresponding to the absorption layer of beta radiation in the concrete. The measurement is very sensitive to the surface contamination but is not sensitive to the contamination in the depth of concrete structure.
Surface contamination monitoring in a survey mode can be used to “hot spot” identification and to demarcation of the areas of increased contamination. Surface contamination monitoring in a fixed positions can be used to the evaluation of the homogeneity of activity distribution on the monitored area.
Ratio of measured surface activity (based on the beta ray detection) and total activity in the structure per unit surface area (based on the gamma ray detection, described in next Chapter) may be an indicator of the contamination depth.

Monitoring of total activity in the structure per unit surface area

An innovative method of *in situ* monitoring of constructional concrete surfaces has been proposed and tested, based on the total activity in the structure per unit surface area (TAUSA) of concrete determination by means of hand-held gamma spectrometer InSpector 1000 with a LaBr detector.

Description of measuring instrument assembly

Hand-held gamma spectrometer InSpector 1000 including multi channel analyzer (MCA) for evaluate 1024 channel energy gamma spectra was used. MCA is equipped with internal accumulator ensuring power supply for 8 hour autonomous operation and internal flash memory for saving acquired energy spectra.

For measurement TAUSA the external scintillation LaBr 1.5” x 1.5” detector (type IPROL-1) equipped with the temperature stabilisation of energy spectra was used. Relative energy resolution is about 4 % at 661.6 keV. A stand for detector fixation in correct counting position can be used.

Description of monitoring method of TAUSA

To evaluate the TAUSA measurements the knowledge of effective contamination depth is required. For most of cases the contaminated surface concrete layer of effective thickness of 1 cm can be supposed. In addition the measurement sensitivity for ^{137}Cs (661.6 keV) does not change significantly in dependence on the presumed thickness of the contaminated surface layer (Figure 4). Due to small thickness of contaminated layer the depth activity distribution near to the exponential dependence can be successfully approximated by even activity distribution.

Detection efficiencies for various effective contamination depths have been determined by a mathematical calibration tool ISOCS based on detector characterisation that was delivered with detector. The characterisation includes the response matrix of point sources in the surrounding space up to distance 500 m from the detector determined on the basis Monte Carlo simulation calculations and verified by empirical measurements.

The even activity distribution in the concrete surface layer and the standard concrete density value 2.35 g/cm^3 have been considered at calculations.

The density of regular grid monitoring network, 1 m, was optimised with respect to the response distribution on a monitored area and to the national legislation requiring the determination of the average values on the surface not larger than 1 m^2. At measurements in small heights above the surface (in contact) at 1 cm contamination depth the effective monitored area is 1 m^2 contributing by about 80% to the total response. The contribution from the farther areas is insignificant. Response distribution on monitored surface in dependence on the contamination depth is introduced in the Table 1, on the Figure 1 and on the Figure 4.

The sensitivity of such measurement of ^{137}Cs at contamination depth of 1 cm was determined to be 3.13 s^{-1}/(Bq/cm^2). The MDA at acquisition time of 100 s and standard external gamma radiation background of 0.1 µSv/h is on the level of 0.5 Bq/cm^2 for ^{137}Cs.

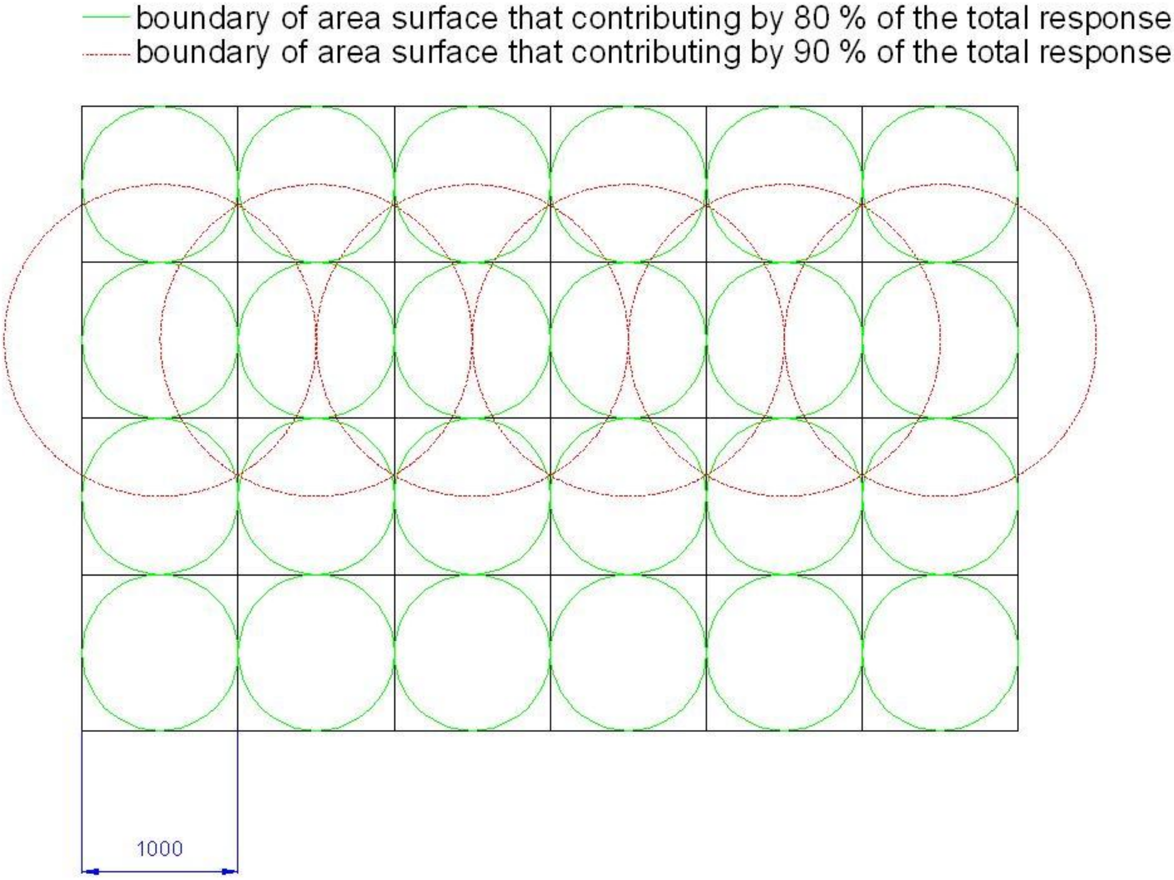
The productivity of monitoring is about 40 m^2 per working shift by two workers including the handling with support equipment.

The assembly was metrologically tested for such measurements by SMÚ Bratislava being classified as activity measuring instrument of radionuclides emitting gamma ray of 15% accuracy class. Testing of calibration method involved tests with surface and point reference sources of various gamma radiation emitting radionuclides posited in various distances from the detector.

Table 1 Dependence of the detected response on contamination depth, at detector placement in the centre of circular concrete surface area

Contamination depth [cm]	Contribution of the response from the circular area with given diameter		
	100 cm	**200 cm**	**400 cm**
0.5	72 %	85 %	92 %
1.0	79 %	89 %	95 %
2.0	85 %	92 %	96 %

Figure 1 Distribution of the detected response on the monitored concrete surface with marked monitoring 1 m regular grid at contamination depth 1 cm



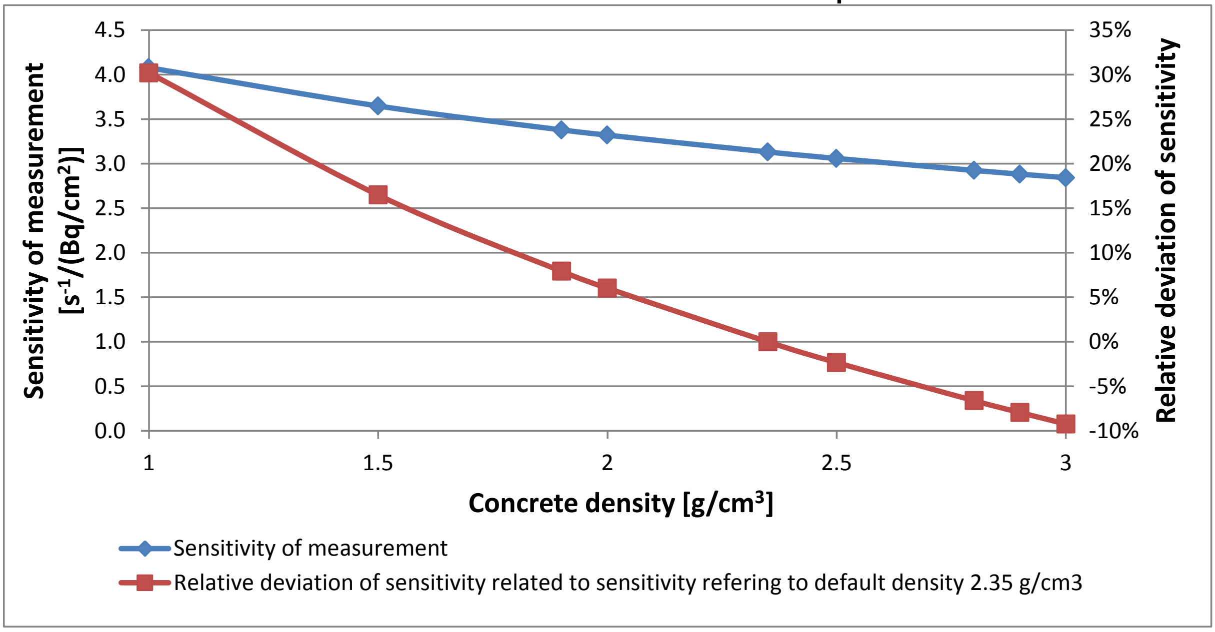
Estimation of average measurement uncertainty

An average measurement uncertainty was evaluated on the base of variation of most significant parameters determining the counting geometry that can affect the result of measurement (the concrete density, detector position, contamination depth, etc.). These parameters are the input data at full energy peak detection efficiency determination by means mentioned ISOCS calibration tool. Uncertainty of detection efficiency calculation by ISOCS tool has been taken to account too. It depends on the energy of radiation and the counting geometry. For most of geometries and energy 661.6 keV it is about 6 %.

Influences of uncertainty of other parameters like counting live time, abundance of gamma ray energy etc. were neglected because they are not important in comparison with parameter described bellow and introduced in the Table 2.

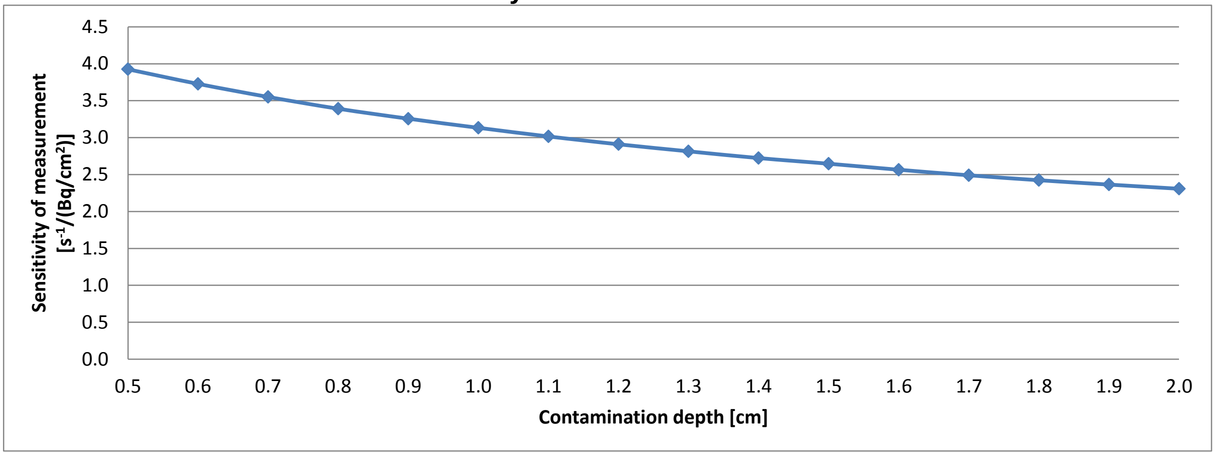
At the detection sensitivity determination the standard value 2.35 g/cm^3 of concrete density with variation ± 0.5 g/cm^3 was supposed. Dependence of the measuring sensitivity on the concrete density is shown on the Figure 2. Influence of material composition is less important than density therefore it was neglected.

Figure 2 Dependence of TAUSA measuring sensitivity on the concrete density of monitored surface for circle area of 4 m diameter and 1 cm contamination depth



Contamination depth in most cases is less than 1 cm. This means thin concrete layer of small account for absorption 661.65 keV gamma ray of ^{137}Cs. If we suppose the accuracy of contamination depth determination in range ± 0.3 cm the uncertainty of detection sensitivity (± 13 %) is not very significant too. Dependence of TAUSA measuring sensitivity on the contamination depth is shown on the Figure 3 as well as on the Figure 4. Supposing of 1 cm contamination depth at TAUSA measurement leads in most cases to conservative approach.

Figure 3 Dependence TAUSA measuring sensitivity on contamination depth, even activity distribution in contaminated concrete layer was considered



The fixed detector position in contact to monitored surface during counting is supposed. In realistic condition the monitored surface is not ideally even due to undulation and small bumps. Effective distance the detector from the monitored surface was considered within range ± 2 cm.

Next significant quantity is net peak area. Uncertainty of its determination was evaluated on the basis of the count of registered events resulting from counting time 100 s and TAUSA threshold limit 8 Bq/cm^2 (see next Chapter).

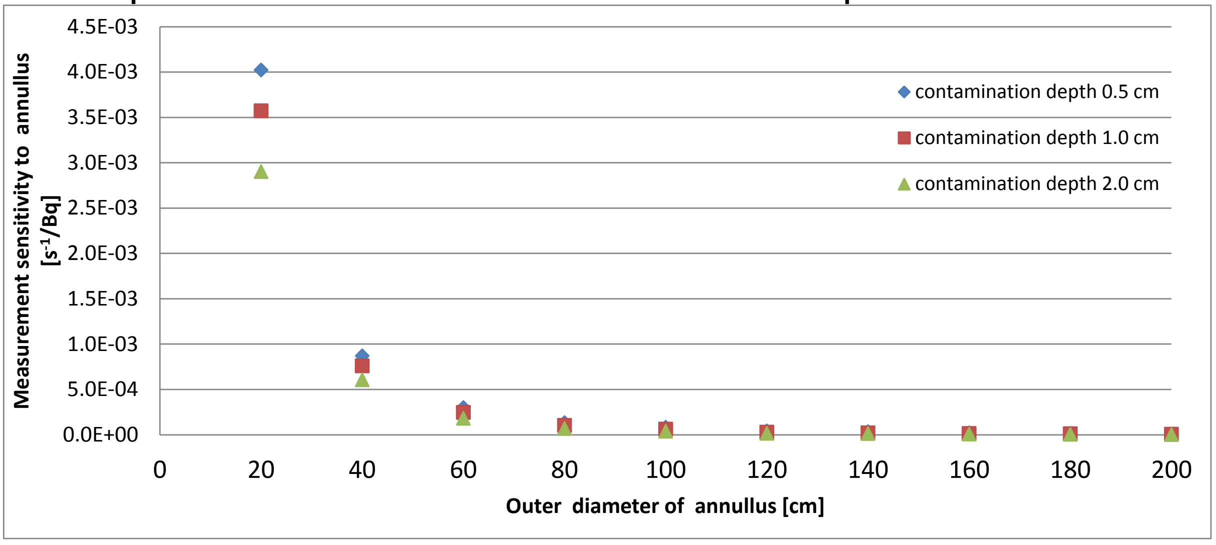
The measurement uncertainty was estimated to be 17 %, provided there is an even activity distribution on the monitored surface (Table 2).

Table 2 Estimation of average uncertainty of TAUSA measurement and its most important components

Source of uncertainty	Uncertainty
Uncertainty of calibration method ISOCS (for 661.6 keV energy)	6%
Uncertainty resulting from change the density of concrete structure (2.35 ± 0.5) g/cm^3	8%
Uncertainty resulting from change the contamination depth (1 ± 0.3) cm	13.3%
Uncertainty resulting from change the detector position during counting (in contact ± 2 cm)	3.7%
Uncertainty of net peak area determination (100 s, As = 8 Bq/cm^2)	3%
Standard combined uncertainty (at homogeneous activity distribution)	**17%**
Uncertainty resulting from inhomogeneous activity distribution on the monitored surface	9%
Standard combined uncertainty (including inhomogeneous activity distribution)	**20%**

The influence of possible uneven activity distribution on monitored surface was also estimated. Maximum measurement sensitivity is reached in the place of the detector position in the centre of the cell of the monitoring grid and with distance from detector is decreased. Distribution of the measurement sensitivity on monitored area is demonstrated on the Figure 4. Let suppose that the monitored area is partitioned by concentric circle lines to particulate annulus, in the centre there is the detector. Measurement sensitivity referring to area of particular annulus is shown on the Figure 4.

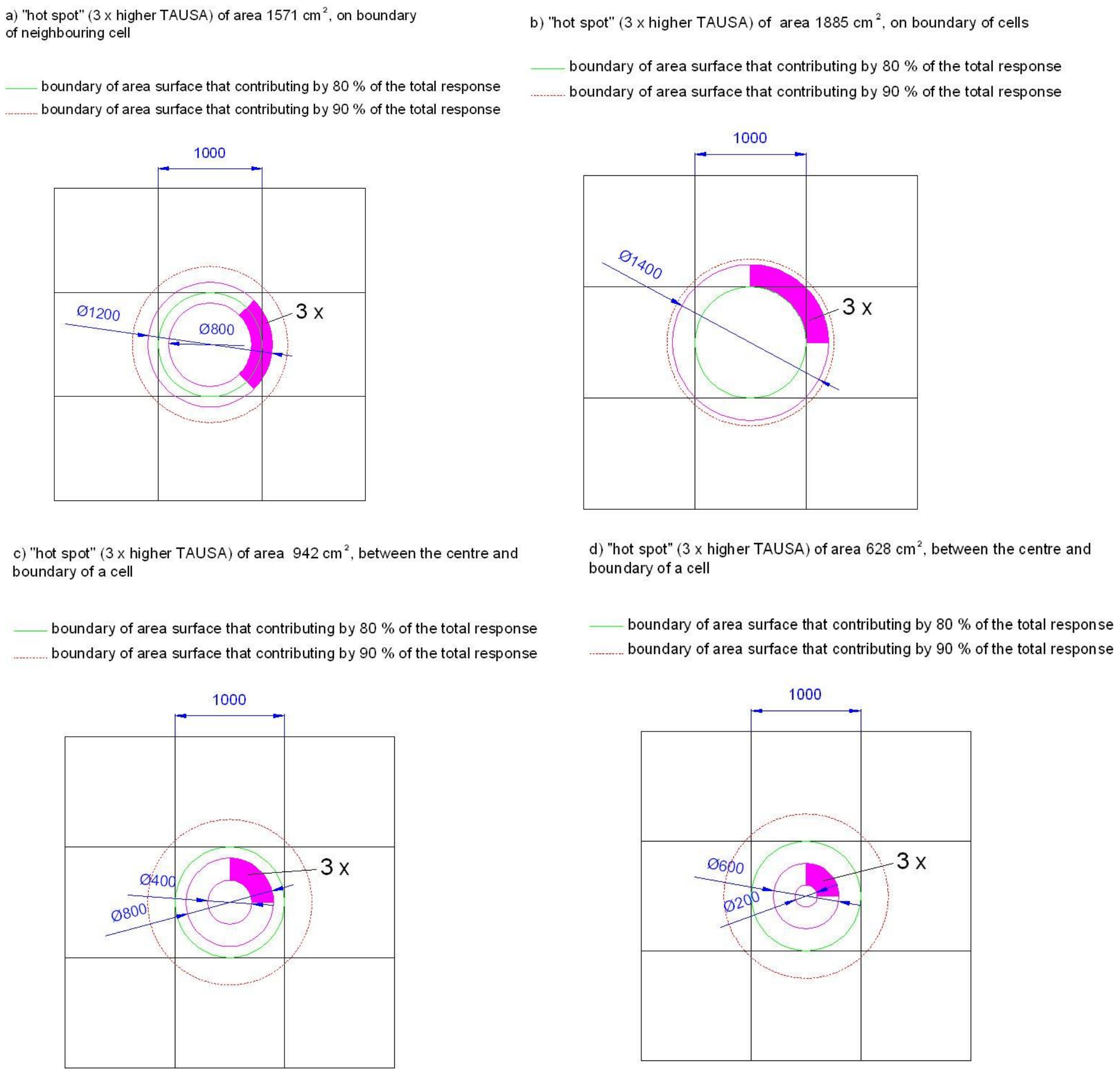
Figure 4 Distribution of measurement sensitivity on monitored area, measurement sensitivity to area of particular annulus is shown at various contamination depths



If we consider even activity distribution on circle surface area 1 m^2 and 3 times higher TAUSA on complement surrounding surface the measurement result will be overestimated about 40 % due to influence from distant places. But this effect will be not significant in neighbours measuring points. Consequently it is reasonable to evaluate only the influence of “hot spots” with small area and in distance less than 1 m from measuring point.

The influence of uneven activity distribution was estimated on the base of different scenarios of presumed "hot spots" occurrence of different dimensions and in different distances from detector. Particular spots were modelled as parts of surfaces with 3 times higher area activity then neighbouring area by sophisticated application of ISOCS calibration tool. Area of particular “hot spots” was about 1000 cm^2 that represent approximately one tenth of effectively monitored area. Scenarios demonstrating “hot spot” occurrence are shown on the Figure 5. Influence of “hot spot” occurrence to measurement result at particular scenarios was evaluated by the deviation of measured activity value (supposing even activity distribution) from real activity value on monitored area (Table 3). Average value of deviations referring to particular scenarios was considered as average uncertainty in consequence unevenly activity distribution on the monitored surface and was estimated to be 9 %. The standard combined uncertainty of measurement, including the possible occurrence of inhomogeneities, was estimated to be 20 %.

Figure 5 Scenarios demonstrating “hot spot” occurrence on the monitored surface area, dimensions are marked in [mm]



e) "Hot spot" (3 x higher TAUSA) of area 1 178 cm^2, boundary of area surface that contributing by 80 % of the total response

boundary of area surface that contributing by 80 % of the total response
boundary of area surface that contributing by 90 % of the total response

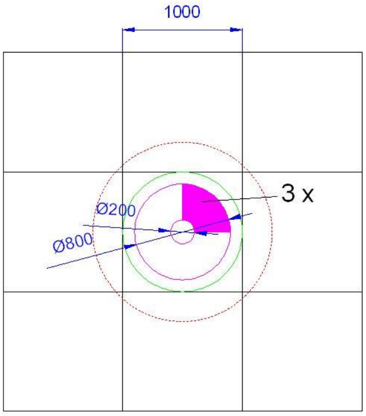


Table 3 Deviations of measured activity value (supposing even activity distribution) from real activity value on the monitored surface area at "hot spot" occurrence according to Figure 5

Scenario	a)	b)	c)	d)	e)	**Average**
Deviation	2 %	0	8 %	16 %	19 %	**9 %**

Results and experience from a pilot in situ monitoring of a large concrete tank

The proposed method of TAUSA *in situ* monitoring was applied at characterisation of a large concrete tank in 2011. This underground tank was used for liquid radioactive waste storage. Tank of cylinder shape with 6 m diameter and 4 m height is split by a mid-line concrete wall to two separately tanks (denoted N1 and N2). Overall area of internal surfaces to be monitored was about 150 m^2.

Average dose rate in the tank was in range 0.2 - 0.5 µSv/h. Increased dose rate level of 7 µSv/h was measured close the pipe lines that were removed firstly.

At the TAUSA measurements the effective thickness of surface contaminated layer 1 cm was supposed. Operative clearance limit 8 Bq/cm^2 for ^{137}Cs was choices on the basis preliminary RN vector for concrete at NPP A1. Average value of TAUSA measurements carry out before decontamination was 8.3 Bq/cm^2, particular values in range from 2 to 70 Bq/cm^2. About 40 % of the total number of the measurements exceeded the level 8 Bq/cm^2. Particular square meters (1 x 1 m^2 cells of monitoring grind) were marked for decontamination. Areas of increased contamination identified by surface contamination survey were marked too. Maps of contamination of tank concrete internal surfaces are on the Figure 6.

For estimation of the effective thickness of the contaminated surface layer the core drill samples were taken, segmented and analysed by laboratory gamma spectrometry. Results of segments analysis have shown that the majority of the contamination is present in the surface layer of thickness < 1 cm. Example of analyse results of core drills are introduced in the Table 4.

The surface concrete layer of thickness 1 up to 5 cm was removed during the decontamination by used hand-held decontamination equipment.

Monitoring of TAUSA was repeated after decontamination. Average value decreased to value 3.7 Bq/cm^2 and all measurements were less than 8 Bq/cm^2.

Samples of the concrete structure were taken to specify hard to detectable radionuclide content by means of radiochemistry analyses and to derivate final clearance level.

Figure 6 Map of the tank concrete internal surface contamination expressed by the total activity in the structure per unit surface area (TAUSA) of ^{137}Cs [Bq/cm^2] measured in the regular grind 1 x 1 m, two single parts of the tank are shown separately

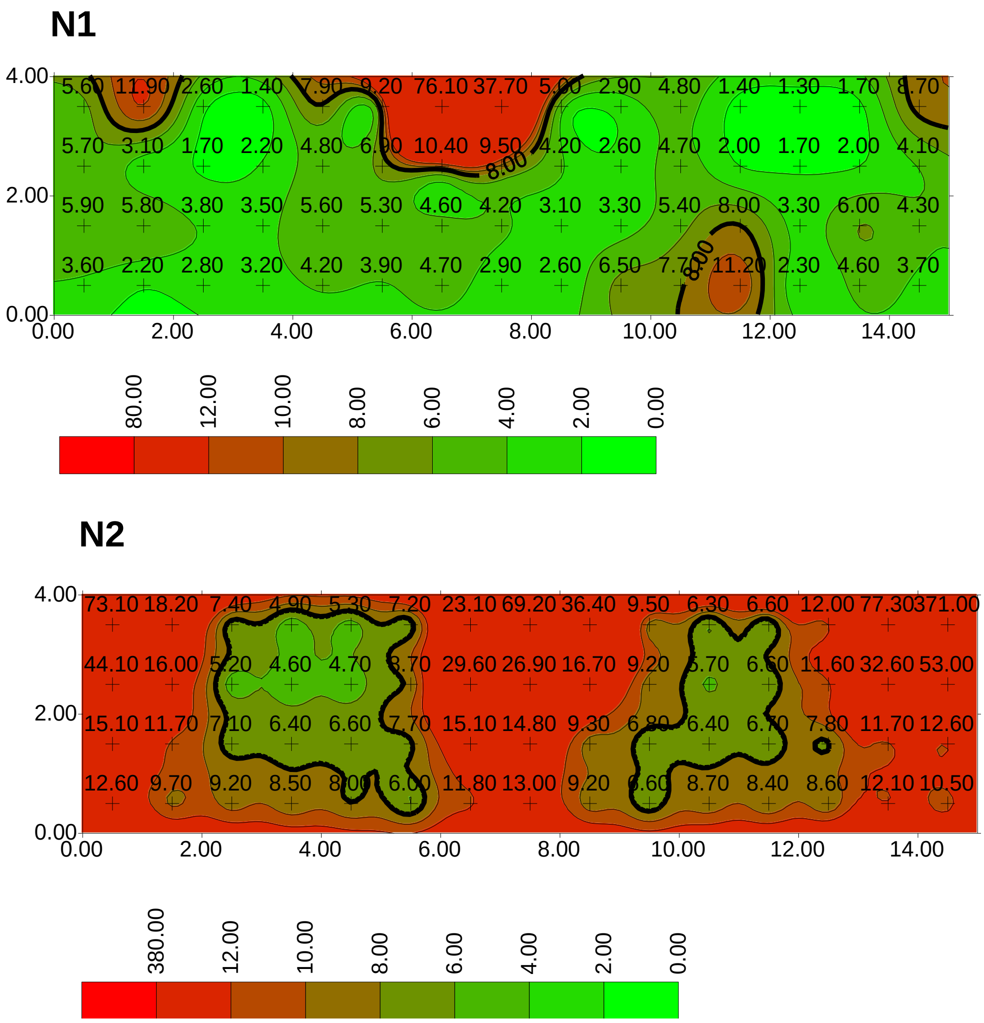


Table 4 Laboratory analysis results of the core drill segments taken from two different places

Sampling point	Core drill	Originating depth of segments [mm]	Massic activity of ^{137}Cs [Bq/kg]
1	1	0 – 10	911
		17 – 55	4.57
	2	0 – 17	602
		17 – 87	<1.23
2	1	0 – 5	1030
		5 – 10	14.7
		10 – 15	9.4
	2	0 – 5	680
		5 – 10	47
		10 – 15	45.3

Conclusions

An innovative method of measurement of the total activity in the structure per unit surface area [Bq/cm^2] that is based on hand-held gamma spectrometer Inspector1000 with LaBr 1.5" x 1.5" scintillation detector was designed and tested. This method is in compliance with international recommendation RP-113. To evaluate the TAUSA measurements the knowledge of effective contamination depth is necessary but for most of cases 1 cm can be supposed.

The monitoring method as well as the other support characterisation methods including dose rate survey, contamination depth determination etc. are described in the paper. Analyse of TAUSA measurement uncertainty was carry out too. Estimated standard combined uncertainty was on the level 17 % at even and 20 % at uneven radioactivity distribution on the monitored area.

The measuring assembly was metrologically tested for such measurements by Slovak metrology institute (SMÚ) in Bratislava and was classified as activity measuring instrument of radionuclides emitting gamma ray of 15% accuracy class.

Introduced approach was tested within decommissioning project of NPP A1 in Slovakia at characterisation of surfaces of the tanks concrete used as liquid radioactive waste storage.

REFERENCE

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