Activity Measurement of Soil Samples Taken in The Vicinity of the RA Research Reactor

Dragana Čekerevac-Mirković, Ivana Maksimović, Miodrag Milošević

Abstract - **During exploitation of the RA heavy water research rector, a high average fuel burnup (7000 MWd/t) was achieved in low enriched (2% of ^{235}U) metal uranium fuel elements. As a result of high fuel burnup and complex fuel element construction (cylindrical shell with aluminium clad), a high leakage of ^{137}Cs in the spent fuel storage pools was detected. According to measurements (in 2012), total activity of ^{137}Cs in spent nuclear fuel storage is assessed to a level of 2 TBq. Until 2008, the ventilation system of spent fuel nuclear storage operated without filters. It was therefore necessary to define the level of the environmental contamination before starting the RA nuclear reactor decommissioning projects; the essential part of this evaluation was determination of the activity of ^{137}Cs and other artificial radionuclides deposited in the soil around the reactor facility. This paper presents the results of radionuclide activity measurement in 44 soil samples taken in the vicinity of the RA and RB reactor buildings, and summarises the comparison with results obtained for control group comprised of 20 samples of uncultivated soil taken from various locations in Serbia. Statistical analysis of these results shown that there is a significant increase of ^{137}Cs activity in the soil around the RA and RB reactor buildings (in average 0.12 Bq/g); on the other side, one should say these values are in the order of values of the clearance level.**

Key words – **The RA nuclear reactor environment, soil samples, Monte Carlo simulation of HPGe detector, non-destructive laboratory measurement.**

I. INTRODUCTION

Exploitation of the RA heavy water research reactor at 6.5 MW, mostly with unalloyed metal uranium fuel elements [1], was accompanied with several peculiarities, in comparison to similar research reactors, which have impact on current decommission process [2]. First, the number of spent fuel elements was very high (6656 fuel elements with unalloyed 2% ^{235}U enriched metal uranium and 1374 with uranium dioxide having initial enrichment of 80% ^{235}U), Ref. [3]. They had a complex construction of fuel in the form of a short tube (100 mm high and 2 mm thick) coated with aluminium (of 1 mm thick) from both sides, giving the total height of 112 mm for one fuel element. The second characteristic, that increases complexity of decommission, is the very high fuel burnup achieved in fuel elements with metal uranium (about 7000 MWd·t^{-1} in average), compared to others nuclear reactors which have used metal uranium (at most 4000 MWd·t^{-1}), Ref. [4]. High fuel burnup implies that the aluminium cladding suffered significant radiation damage, which resulted in impaired fuel integrity during cooling phase in the spent fuel storage. Finally, although the RA nuclear reactor had no accident with core damage, there have been several incidents - twowith nuclear fuel, two with a leak of heavy water, and few incidents with contamination of the primary circuit and the reactor building. An overview of these events is presented in the Ref. [5]. Consequently, there was a high leakage of ^{137}Cs and ^{90}Sr in the water of spent fuel storage pools. In 2012, two years after the repackaging and transporting of spent fuel elements in the country of origin (Russia), the reactor research group performed in situ radiological characterisation of four ion-exchanger columns used for removal of ^{137}Cs from contaminated water in these pools. It was found, that the activity of caesium which had been removed was about 0.9 TBq. It is estimated that the remaining activity of ^{137}Cs in the long open stainless steel containers is at least as high, which means that approximately 2 TBq of ^{137}Cs has been leaked from spent fuel elements.

The activity increase of radionuclides in the water pools lead to an increase of activity of radioactive aerosols released in the spent fuel storage rooms, due to the water evaporation [3]. Before installation of the mobile ventilation unit PFB-2500 HEPA, with the efficiency of 99.97% for the removal of aerosols, the spent storage room was vented only with the ventilation system (V3), without HEPA filtration. Thus, until the 2008, when HEPA ventilation was installed, there was no barrier for the release of radionuclide ^{137}Cs and other artificial radionuclides into the environment, and their consequent deposition in the soil around the RA nuclear rector buildings.

This paper presents the results of radionuclide activity measurement in soil samples taken in the vicinity of the RA and RB reactor buildings, and in samples of uncultivated soil taken from various locations in Serbia.

II. MATERIAL AND METHODS

For the purpose of assessing the need for remediation of soil on the site of the RA and RB nuclear reactors, it was necessary to measure radionuclide activity in soil samples taken from depth, taking into account that the soil type influences the rate of caesium migration downward. It was assumed that the rate of vertical migration of caesium for the Danube type of soil is around 1 cm a year, and the sampling depth of 15 cm is chosen having in mind that there was no caesium deposition after installing ventilation system with HEPA filters (in 2008).

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All soil samples were collected in standard plastic containers, filled to the top, with volume of 137.4 cm^3. No chemical or mechanical treatment of samples was performed, in order to ensure reliable activity measurement of indirectly measured radionuclides. Figure 1 illustrates non-destructive sampling procedure from the depth, denoted by *d*. This sampling procedure excludes any physical or chemical actions that could disturb the existing equilibrium between radionuclides.

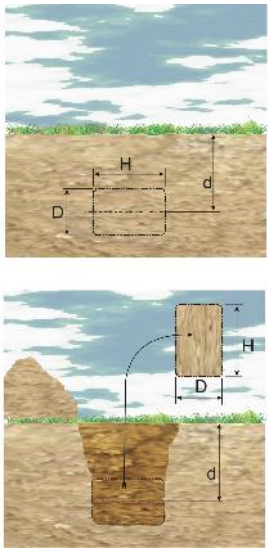

Fig. 1. Sampling from the depth d

Around the RA nuclear reactor building 22 samples were taken. Additionally, around the building of the RB nuclear reactor 10 samples were collected, and another 12 samples outside the fence surrounding the reactors site. Figures 2 and 3 show sampling locations around the buildings of the RA and RB reactors, respectively, while figure 4 presents sampling locations on the wider area outside the reactor site fence.

The control group comprises 20 samples of uncultivated soil taken on random locations in Serbia, excavated on the same way as the soil samples at the RA and RB reactor site. Results show that in some cases there is a large difference between the activity measurements in soil taken from the reactor site and those ones from nationwide average levels.

Fig. 2. Sample locations in the vicinity of the reactor RA building

Fig. 3. Sample locations in the vicinity of the reactor RB building

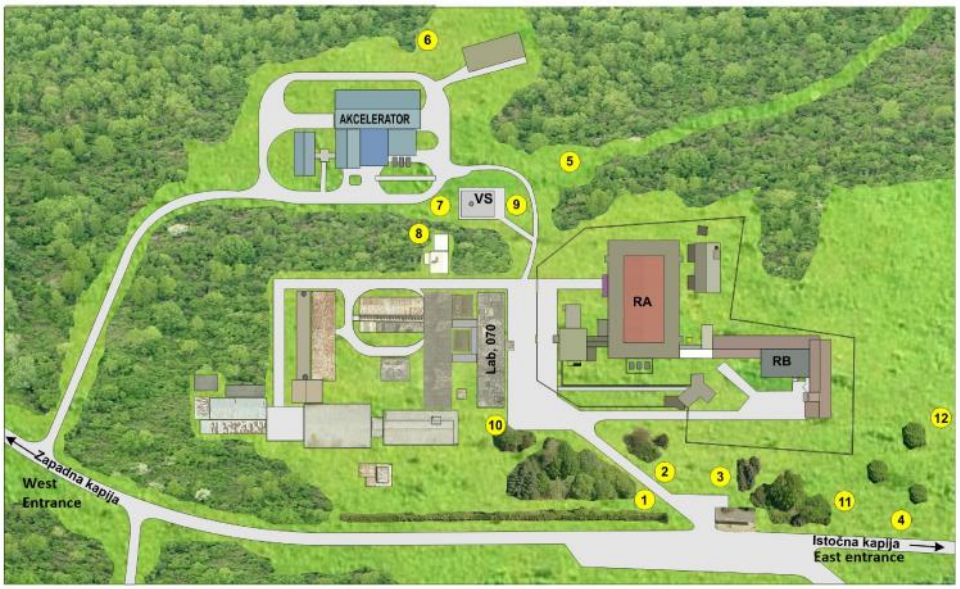

Fig. 4. Sample locations outside the reactor site fence

Activity of these samples were measured using coaxial HPGe detector with Be window, the GX5020 detector manufactured by Canberra (Meriden, Conectitat), connected to a Multiport II multichannel analyser that uses the Genie-2000 gamma analysis software [6]. The detector was specified as having a 50% efficiency relative to a 3x3″ NaI(Tl) detector at 1.33MeV.

Due to a large number of samples with various soil densities, there was a need to automated processing of results. For this purpose, the SOILACT code, written in FORTRAN and described in Ref. [7] was used. In this code the activity (A) of identified radionuclide was determined in standard manner using characteristic gamma rays,

$$A = \frac{C}{\varepsilon \, Y \, COI}, \qquad (1)$$

i.e., using measured peak area for chosen lines of gamma radiation (C), yield (Y), numerically determined detector efficiency (ε) and numerically determined factor of coincidence summing effects for Ge detector (COI factor). Numerical determination of the efficiency and COI factor (when coincidence summing of gamma and X-rays exists) for the GX5020 detector and soil sample (of 137.4 cm^3) is based on a geometry model of this detector, described in Ref. [8]. This model was developed for the Monte Carlo program MCNP-5 [9] in the Department for Development and Application of Nuclear Technologies of the PC NFS. The model was adjusted by optimising dead layer thickness of used detector so that the simulated efficiencies match the experimentally found values. It was shown (Ref. [8]) that for all standard sources, (point sources, cylindrical and Marinelli beakers), angles and energies (10 keV and 2000 keV) tested, the differences between measured and Monte Carlo simulated efficiency were less than specified uncertainties of used standards sources, i.e., less than 4%.

The efficiency of the GX5020 detector for non-destructive prepared soil samples is very sensitive on soil composition (volume fractions of dry soil, water and air), particularly for low energy (less than 200 keV) gamma and X-rays. For this reason, the SOILACT code uses the detector characterisation, in which the values of ε and COI parameters were calculated with the MCNP-5 code as function of two variable: the density of soil (ρ) and volume fraction of dry soil in sample (v_S). In this characterisation process, the volume fraction of water (v_W) in soil sample was obtained by preserving the measured soil sample density (ρ)

$$\rho = \rho_S v_S + \rho_W v_W + \rho_{air}(1 - v_S - v_W), \qquad (2)$$

where ρ_S, ρ_W and ρ_{air} are dry soil, water and dry air densities, equal to 1.9 $g \cdot cm^{-3}$, 1 $g \cdot cm^{-3}$ and 0.00125 $g \cdot cm^{-3}$, respectively. The composition of dry soil was taken from Ref. [10]. For each soil sample density (i.e. for each weight of soil sample in the range from 60 g to 260 g), ε and COI parameters are calculated for three values of volume fraction of dry soil in sample $v_{S,min}$, $v_{S,av}$ and $v_{S,max}$. The minimum and maximum values $v_{S,min}$ and $v_{S,max}$ were obtained for sample containing only water and dry soil (without air), and only dry soil and dry air (without water), respectively. The $v_{S,av}$ represents the average value of volume fraction of dry soil

$$v_{av} = 0.5 \cdot (v_{S,min} + v_{S,max}) \qquad (3)$$

The dependence of ^{137}Cs efficiency on soil sample weight calculated for average volume fraction of dry soil in sample is given in figure 5.

Due to presented discretization of volume fraction of dry soil, the SOILACT code is applicable only for samples containing dry soil, water and air. The procedure for activity measurement of soil samples with significant quantities of metal scraps, uranium or thorium (with weighting fraction of few percents) is given in Ref. [11].

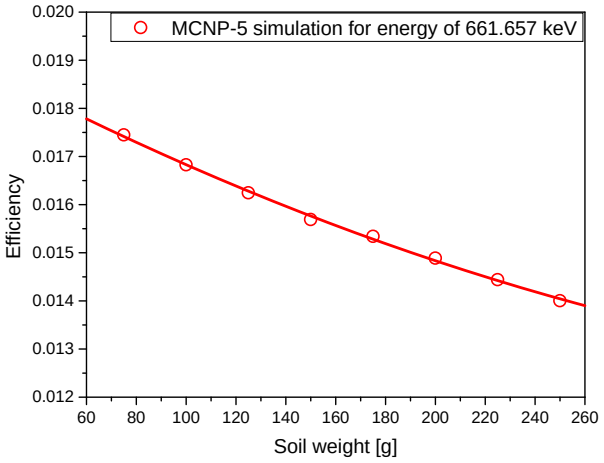

Fig. 5. Efficiency for ^{137}Cs as function of soil weight in sample calculated for average volume fraction of dry soil in sample

For each soil sample with density ρ, the SOILACT code starts analysis with ε and COI parameters calculated for average volume fraction of dry soil $v_{s,av}$. In cases when sample contains radionuclide with several gamma lines, the SOILACT code allows the estimation of the soil sample composition. This is done by selecting the volume fraction of dry soil (v) in a way that the activity calculated for the different gamma lines of the same radionuclide (in this paper it is ^{238}U) are similar (i.e. the differences are less than 7%).

In order to avoid errors due to the net area calculations, the measured pulse height distributions (PHD) obtained with the GX5020 detector and Monte Carlo simulated PHDs for this detector, were analysed using the ANGES software [12]. This software allows a selection of optimal parameters in the Gaussian fitting procedure of peaks, which is very important for multiple peaks in cases of low total counts (as is the case with the ^{241}Am and ^{234}Th peaks at energies 59.54 keV and 63.29 keV in the environmental samples).

The measurement of uncertainties was performed according to the ISO/BIMP ″Guide to the Expression of Uncertainty in Measurement″, taking into account the statistical uncertainty of the measured peak area, as well as efficiency calibration uncertainty.

III. Results

Analysis of the energy spectra of gamma radiation from soil samples taken in vicinity of the RA reactor, revealed several radionuclides: ^{238}U, ^{232}Th, ^{226}Ra, ^{241}Am, and ^{137}Cs. Characteristic gamma lines used in the code SOILACT to determine activity of these radionuclides in soil samples are presented in table I. These data are taken from the NuDat2.6 evaluation [13].

The results obtained with the SOILACT code for 22 soil samples taken around the RA nuclear reactor building (figure 2) are shown in table II. Table III contains the results obtained with this code for 10 soil samples taken in the vicinity of the RB nuclear reactor building (figure 3). The results obtained on the same way for 12 soil samples taken outside the site fence (figure 4) are presented in tables IV. The results of specific activity measurements performed with presented procedure for 20 soil samples of the control group are given in table V.

TABLE I
Characteristic gamma lines used to determine activity of radionuclides in soil samples

Radionuclide	Gamma emitter	Energy [keV]	Yield [%]
^{238}U	^{234}Th	63.290	3.72
	^{234}Th	92.590	4.25
^{232}Th	^{228}Ac	911.204	25.80
	^{228}Ac	968.971	15.80
^{226}Ra	^{226}Ra	186.211	3.64
^{241}Am	^{241}Am	59.537	35.90
^{137}Cs	^{137m}Ba	661.660	85.10
^{40}K	^{40}K	1460.8220	10.66

TABLE II
Results for soil samples taken around RA building

No.	Specific activity [Bq·g^{-1}]		
	^{232}Th	^{238}U	^{137}Cs
1	0.033 ± 0.002	0.017 ± 0.001	0.238 ± 0.012
2	0.033 ± 0.002	0.024 ± 0.002	0.027 ± 0.002
3	0.033 ± 0.002	0.017 ± 0.001	0.316 ± 0.014
4	0.013 ± 0.001	0.010 ± 0.001	0.099 ± 0.005
5	0.037 ± 0.002	0.026 ± 0.002	0.153 ± 0.007
6	0.041 ± 0.002	0.022 ± 0.002	0.107 ± 0.005
7	0.034 ± 0.003	0.029 ± 0.002	0.020 ± 0.001
8	0.040 ± 0.002	0.022 ± 0.001	0.049 ± 0.002
9	0.034 ± 0.001	0.019 ± 0.001	0.105 ± 0.004
10	0.042 ± 0.002	0.027 ± 0.002	0.021 ± 0.001
11	0.036 ± 0.001	0.025 ± 0.001	0.188 ± 0.008
12	0.034 ± 0.001	0.022 ± 0.001	0.112 ± 0.005
13	0.040 ± 0.002	0.026 ± 0.002	0.018 ± 0.001
14	0.010 ± 0.001	0.008 ± 0.001	0.180 ± 0.008
15	0.037 ± 0.001	0.019 ± 0.001	0.364 ± 0.015
16	0.043 ± 0.002	0.028 ± 0.002	0.138 ± 0.006
17	0.039 ± 0.002	0.025 ± 0.002	0.180 ± 0.008
18	0.036 ± 0.002	0.022 ± 0.001	0.150± 0.006
19	0.046 ± 0.002	0.024 ± 0.002	0.071 ± 0.003
20	0.039 ± 0.002	0.028 ± 0.002	0.086 ± 0.004
21	0.039 ± 0.002	0.023 ± 0.002	0.348 ± 0.014
22	0.039 ± 0.002	0.024 ± 0.002	0.128 ± 0.005

TABLE III
RESULTS FOR SOIL SAMPLES TAKEN AROUND RB BUILDING

No.	Specific activity [Bq·g^{-1}]		
	^{232}Th	^{238}U	^{137}Cs
1	0.033 ± 0.002	0.017 ± 0.001	0.238 ± 0.012
2	0.033 ± 0.002	0.024 ± 0.002	0.027 ± 0.002
3	0.033 ± 0.002	0.017 ± 0.001	0.316 ± 0.014
4	0.013 ± 0.001	0.010 ± 0.001	0.099 ± 0.005
5	0.037 ± 0.002	0.026 ± 0.002	0.153 ± 0.007
6	0.041 ± 0.002	0.022 ± 0.002	0.107 ± 0.005
7	0.034 ± 0.003	0.029 ± 0.002	0.020 ± 0.001
8	0.040 ± 0.002	0.022 ± 0.001	0.049 ± 0.002
9	0.034 ± 0.001	0.019 ± 0.001	0.105 ± 0.004
10	0.042 ± 0.002	0.027 ± 0.002	0.021 ± 0.001

TABLE IV
RESULTS FOR SOIL SAMPLES TAKEN OUTSIDE REACTORS SIDE FENCE

No.	Specific activity [Bq·g^{-1}]			
	^{232}Th	^{238}U	^{241}Am	^{137}Cs
1	0.050 ± 0.002	0.036 ± 0.002	<MDA (0.0005)	0.022 ± 0.001
2	0.050 ± 0.002	0.042 ± 0.002	<MDA (0.0005)	0.030 ± 0.001
3	0.048 ± 0.002	0.039 ± 0.002	0.0013 ± 0.0004	0.066 ± 0.003
4	0.049 ± 0.002	0.039 ± 0.002	0.0009 ± 0.0004	0.050 ± 0.002
5	0.019 ± 0.001	0.019 ± 0.001	0.0017 ± 0.0003	0.105 ± 0.004
6	0.047 ± 0.002	0.031 ± 0.002	0.0024 ± 0.0005	0.025 ± 0.001
7	0.042 ± 0.002	0.031 ± 0.002	0.0017 ± 0.0004	0.059 ± 0.003
8	0.042 ± 0.002	0.027 ± 0.002	0.0021 ± 0.0005	0.005 ± 0.001
9	0.045 ± 0.002	0.032 ± 0.002	0.0014 ± 0.0005	0.047 ± 0.002
10	0.040 ± 0.002	0.031 ± 0.002	0.0007 ± 0.0004	0.064 ± 0.003
11	0.048 ± 0.002	0.036 ± 0.002	0.0009 ± 0.0004	0.043 ± 0.002
12	0.044 ± 0.002	0.032 ± 0.002	0.0008 ± 0.0003	0.032 ± 0.001

TABLE V
RESULTS FOR SOIL SAMPLES OF CONTROL GROUP

Location	Specific activity [Bq·g^{-1}]		
	^{232}Th	^{238}U	^{137}Cs
Dolovo 1 (Pančevo)	0.036±0.002	0.032±0.002	0.023±0.001
Dolovo 2 (Pančevo)	0.032±0.001	0.024±0.001	0.016±0.001
Fruška Gora	0.030±0.002	0.020±0.001	0.012±0.001
Jajinci 1 (Beograd)	0.048±0.002	0.037±0.002	0.069±0.003
Jajinci 2 (Beograd)	0.047±0.002	0.036±0.002	0.031±0.001
Jajinci 3 (Beograd)	0.045±0.002	0.037±0.002	0.049±0.002
Dorćol 1 (Beograd)	0.068±0.002	0.023±0.001	0.006±0.001
Dorćol 2 (Beograd)	0.037±0.002	0.022±0.001	0.013±0.001
Kraljevo	0.038±0.002	0.034±0.002	0.029±0.001
Kremna (Užice)	0.013±0.001	0.010±0.001	0.019±0.001
Kruševac	0.036±0.002	0.025±0.002	0.035±0.002
Ljig	0.046±0.002	0.037±0.002	0.006±0.001
Ljubić (Čačak)	0.048±0.002	0.031±0.002	0.023±0.001
Novi Sad	0.030±0.002	0.023±0.002	0.006±0.001
Pančevo	0.028±0.002	0.028±0.002	0.033±0.002
Smederevo	0.038±0.002	0.028±0.002	0.015±0.001
Sremska Kamenica	0.048±0.002	0.036±0.002	0.022±0.001
Zlatari 1 (Kruševac)	0.033±0.002	0.021±0.001	0.007±0.001
Zlatari 2 (Kruševac)	0.035±0.002	0.031±0.002	0.004±0.001
Zlatari 3 (Kruševac)	0.044±0.002	0.031±0.002	0.017±0.001

The analysis of obtained results, based on Student's *t*-test, shown that there is no statistically significant difference between ^{238}U specific activities in soil samples taken from the RA and RB nuclear reactors site (total of 44 samples) and soil samples in control group (total of 20 samples). The average values of ^{238}U activity in these two sets of soil samples are 0.027 Bq·g^{-1} and 0.028 Bq·g^{-1}, respectively. These average values are very similar to ^{238}U specific activity in the Earth's crust, equal to 0.033 Bq·g^{-1}, according to Ref. [14]. The same test shown that there is no statistically significant difference between ^{232}Th specific activities in soil samples taken from the RA and RB nuclear reactors site and soil samples in control group. The average values of ^{232}Th specific activity in

these two sets of samples, equal to 0.038 $Bq \cdot g^{-1}$ and 0.039 $Bq \cdot g^{-1}$, respectively, are very similar to ^{232}Th specific activity in the Earth's crust, specified as 0.045 $Bq \cdot g^{-1}$ in Ref. [14]. On the same way, it was found that average values of ^{226}Ra specific activity in these two set of soil samples, equal to 0.038 $Bq \cdot g^{-1}$ and 0.040 $Bq \cdot g^{-1}$, respectively, are very similar to ^{238}U specific activity in the Earth's crust. These findings confirm that there are no ^{238}U, ^{226}Ra and ^{232}Th of artificial origin at the RA and RB nuclear reactors site.

The artificial actinide ^{241}Am, was found only in the samples taken outside the RA and RB nuclear reactors site fence (figure 4, and table IV). Maximum measured specific activity of ^{241}Am (0.0024 ± 0.0005 $Bq \cdot g^{-1}$) was less than *clearance level*, equal to 0.100 $Bq \cdot g^{-1}$ according to domestic rulebook [15], and European Commission (EC) Directive [16] and IAEA Guidance [17].

The second artificial radionuclide found in all measured soil samples was ^{137}Cs. The analysis of results obtained for ^{137}Cs (based on Student's *t*-test) shown that there is a statistically significant difference between specific activities in soil samples taken from the RA and RB nuclear reactors site (total of 44 samples) and soil samples in control group (total of 20 samples). The average values of ^{137}Cs specific activity in these two sets of soil samples are 0.119 $Bq \cdot g^{-1}$ and 0.022 $Bq \cdot g^{-1}$, respectively. The greater average value of ^{137}Cs activity in first set of soil samples (by a factor of 5.5) confirms a small leakage of ^{137}Cs in the immediate surroundings of the RA nuclear reactor in the past. It should be noted that the maximum measured specific activity of ^{137}Cs (0.364 ± 0.015 $Bq \cdot g^{-1}$) was less than *clearance level* value (equal to 1.000 $Bq \cdot g^{-1}$) in the domestic rulebook [15], and was greater than *clearance level* value given in the EC Directive [16] and IAEA Guidance [17] (equal to 0.100 $Bq \cdot g^{-1}$). However, the average value of ^{137}Cs specific activity in soil samples taken from the RA and RB nuclear reactors site (0.119 $Bq \cdot g^{-1}$) is very close to the *clearance level* value for ^{137}Cs, recommend in the most recent EC and the IAEA recommendations [16,17].

IV. Conclusion

This study confirmed the previous expectations about the moderate release of ^{137}Cs into the environment of the reactor RA in the past. Nevertheless, the specific activity of ^{137}Cs in soil around the reactor RA is very close to general clearance level established by the international standards. Therefore, further surveys are needed in order to determine the vertical profile of ^{137}Cs deposition. The final conclusion on the soil contamination cannot be drawn from these results only, due to a typical heterogeneous spread of ^{137}Cs in the environment. However, these results are important since they provide an insight on the state of contamination in the superficial soil layer, prior to further stages of reactor RA decommission.

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