

SOIL ACTIVITY LEVELS IN THE DECOMMISSIONING AREA OF A VVR-S NUCLEAR RESEARCH REACTOR

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Abstract. The paper presents the methodology for evaluating the levels of soil activity in the area possibly affected by decommissioning activities of the VVR-S Nuclear Research Reactor from National Institute for Research and Development in Nuclear Physics and Engineering, Romania. Gamma spectrometric analyses of the soil samples sampled from the surface and depth were carried out in order to obtain information about possible recent contamination from radioactive dust in the air as well as from unknown past "accidents". The analysis was performed both before reactor decommissioning began (year 2006) and after its completion (year 2020) to demonstrate that the non-restrictive release criteria under the regulatory body regime for land from decommissioning area are met. Activity levels of anthropogenic and natural radionuclides do not exceed the exclusion levels imposed by the basic domestic safety standards in force. It can be stated that the reactor operation and decommissioning did not generate contamination of the surrounding soil.

Key words: soil contamination, reactor, decommissioning

1. INTRODUCTION

The VVR-S type nuclear research reactor with thermal neutrons from IFIN-HH was the first one of this type put into operation in the South-East of Europe. The main purpose was radioisotopes production for medical and industrial applications and research in physics, biophysics and biochemistry. Reactor was operated between 1957 and 1997 at a nominal thermal power of 2 MW, using low-enriched (10%) EK-10 type and high-enriched (36%) S-36 type as nuclear fuel and cooled and moderate with distillate water [1]. In 1997 the reactor operation was stopped and it was put under conservation, until 2002. Due to safety reasons, the reactor was legally permanently shut-down in 2002 and decommissioned between 2010-2020. The

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decommissioning area (land and buildings) taken into considering in our study is presented in Figure 1.

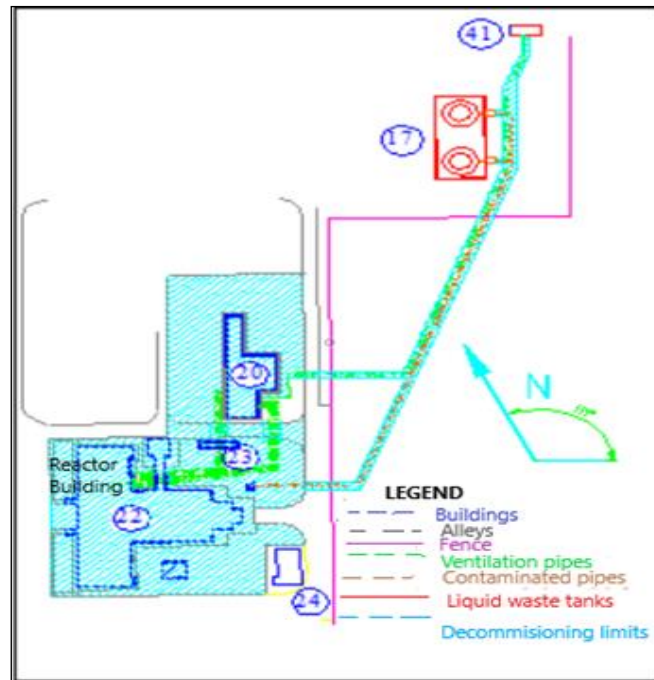


Fig. 1. VVR-S Nuclear Research Reactor decommissioning area

The reactor internal and external systems were contaminated during operation due to the activated corrosion and fission products transport and deposition (leakages). The main reactor components and systems such as the Reactor Block, Cooling Pond for Nuclear Spent Fuel Assemblies, Hot Cells for the irradiated materials processing, Radioactive Liquid Effluents Purifying System, Collecting and Drainage Systems, the Underground Buffer Tank (30 m³), the Primary and Secondary Cooling Circuit, De-gasser System, the Ventilation System, were located in the Reactor Building. A Spent Nuclear Fuel Storage facility (SNFS) was also under operation near the Reactor Building. The spent fuel was stored in the distillate water, inside of the SNFS' pools. Nowadays, this facility is used for aluminium and graphite wastes interim storage resulted from reactor decommissioning.

The main potential sources of the soil pollution during reactor operation and decommissioning were the aerosols and gases released into the atmosphere through the reactor chimney, the radioactive liquid effluents accidentally discharged as well as the solid waste stored uncontrolled on the soil surface [1]. During the reactor operation and decommissioning, a ventilation system with air filters was used to

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evacuate and reduce radioactive contamination from the air in the technical areas of the reactor. Also, to prevent environmental pollution, the radioactive gaseous effluents were monitored and discharged into the atmosphere through a 40 m high chimney equipped with high-resolution HEPA filters [1].

The radioactive liquid effluents were collected in a controlled manner to prevent soil contamination. Thus, after generation there were transferred into the stainless-steel underground buffer tank (30 m³ capacity) and then into the 300 m³ tanks at the radioactive waste treatment plant through the stainless-steel underground pipe system. The underground pipes were decommissioned in 2015 and the buffer tank in 2018.

2. SOIL INVESTIGATION METHODOLOGY

In order to detect the degree of soil pollution around the reactor as a result of its operation, a number of 182 soil samples were sampled and analysed between 2005-2006. Then, at the end of the reactor decommissioning in 2020, 60 samples were sampled and analysed in the same manner in order to detect potential contamination as a result of reactor decommissioning.

The samples were sampled both from the soil surface (10 cm) and depth (30 cm), from the points located in the reactor surrounding area and analysed by gamma ray spectrometry method to determine the activity levels of the anthropogenic and naturally occurring radionuclides. The soil surface was investigated to obtain the information about the contamination caused by the recent airborne dust. Soil samples from in-depth were investigated to detect the contamination over a longer period caused by the past "unknown" accidents or those may reflect the natural background level.

The collection and analysis of soil samples between 2005 and 2006, was carried out by an external expert entity, the National Institute for Research and Development in Metals and Radioactive Resources (INCDMR). The aim was to achieve the "Environmental Assessment for the decommissioning of the VVR-S Nuclear Research Reactor". For this purpose, the samples were taken from the reactor's surroundings from an area of approximately 10,000 m², on parcels of 10 m x 10 m, according to the investigation plan (see Figure 2) [2]. Particular attention was paid to the route of the radioactive effluents pipeline.

The methodology for soil sampling consisted in the following steps: the surface around each sampling point was cleaned of branches, soil and vegetation, then 250 g soil were put in a plastic bag to be prepared for gamma ray spectrometry measurement. Depth samples (30 cm) were collected using a core drill (see Figure 2) [2].

At the end of reactor decommissioning in 2020, the soil sampling and analysing was performed by the IFIN-HH Reactor Decommissioning Department in order to prepare the “Environmental Assessment Report at the end of decommissioning”. Samples were collected on 3,000 m² surface from reactor decommissioning area (see the Figure 3), using the same methodology as it 2006. The radioactive effluent pipeline route was not investigated due to was decommissioned in 2015 and the analyses revealed that the soil was not contaminated. Also, the area around the former SNFS was not investigated due to the building changed its destination to a Solid Radioactive Waste Interim Storage, currently under the IFIN-HH operation and surveillance.

The soil activity levels from the years 2006 and 2020 are compared to demonstrate that the soil is not contaminated due to reactor operation and decommissioning and the free release criteria under Regulatory Body regime are met at the end of decommissioning.

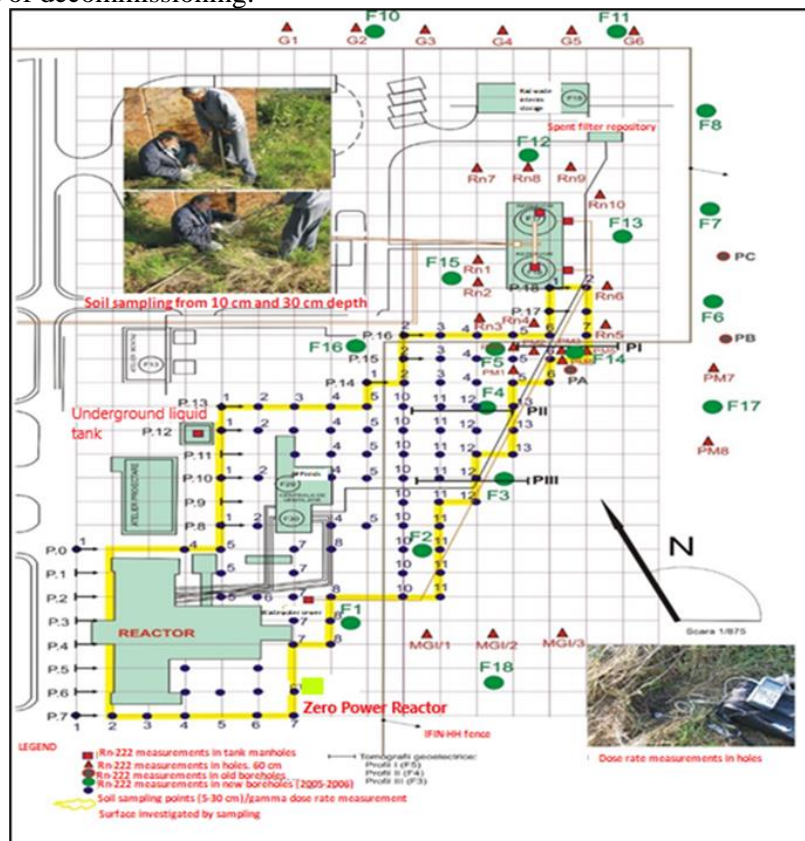


Fig. 2. Soil investigation plan at the end of Reactor operation [2]

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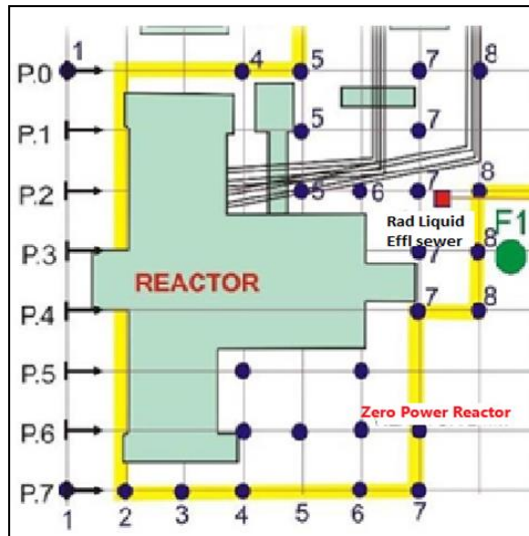


Fig. 3: Soil investigation plan at the end of Reactor decommissioning

3. METHODOLOGY FOR RADIONUCLIDE MEASUREMENTS

3.1 SOIL SAMPLES PREPARATION

The samples were prepared in laboratory for gamma spectrometry measurements according to the specific procedure. Thus, were put onto metallic trays, dried in oven at 105°C to reach a constant mass, crushed and sieved ($\Phi < 2$ mm) to remove impurities (gravel, wood, bricks, concrete), weighed and transferred into cylindrical plastic boxes. The boxes were closed tightly and left for 30 days to achieve the radioactive equilibrium between ^{226}Ra and its descendants ^{214}Pb and ^{214}Bi and then, measured by the gamma ray spectrometry method [3].

3.2 GAMMA RAY SPECTROMETRY MEASUREMENTS

The type and activity levels of the radionuclide components were determined by the notified laboratories of the research institutes INCDMR and IFIN-HH. In 2006, the analyses were performed by INCDMR using the ACCUSPEC high resolution gamma spectrometry system equipped with a high-resolution semiconductor detector HPGe CANBERA type GC 2018-7500 SI.

In 2020, samples were analysed by Radioactive Waste Management Department (RWMD) Laboratory for Radionuclide, Physics-Chemical, Mechanical

and Structural Characterization (LRPMSC) using a high-resolution gamma spectrometry system type CANBERA, with DSA 1000 multichannel analyser, HPGe high resolution semiconductor detector, 30% efficiency, energy range 50 – 2000 KeV, according with the specific procedure [3].

4. RESULTS

The preliminary radiological characterisation of the reactor block components carried out before decommissioning [4] as well as the gamma spectrometry analyses of the radioactive waste drums resulting from the cutting, disassembling and clean-up of reactor performed during the decommissioning process [5], revealed the presence of the following anthropogenic radionuclides: ^{22}Na , ^{60}Co , ^{94}Nb , ^{106}Ru , ^{105}Sb , ^{129}I , ^{133}Ba , ^{134}Cs , ^{137}Cs , ^{152}Eu , ^{154}Eu , ^{155}Eu , $^{166\text{m}}\text{Ho}$, ^{235}U and ^{241}Am . The Minimum Detectable Activities (MDA) for the radionuclides of interest are presented in Table 1.

Table 1.

Minimum Detectable Activities [Bq/kg] for the radionuclides of interest

	^{22}Na	^{60}Co	^{94}Nb	^{106}Ru	^{105}Sb	^{129}I	^{133}Ba	^{134}Cs	^{137}Cs	^{152}Eu	^{154}Eu	^{154}Eu	$^{166\text{m}}\text{Ho}$	^{235}U	^{241}Am
min	1.1	0.8	0.9	8.6	2.9	0.0	1.7	1.2	0.7	2.2	3.1	1.8	1.1	9.9	2.3
max	2.8	2.7	2.2	20.8	6.5	428.0	3.5	2.8	2.6	4.8	8.0	4.5	2.4	25.5	9.2

Gamma spectrometric analyses of the soil samples performed at the end of reactor operation (in 2006) and decommissioning (in 2020) revealed the presence of both anthropogenic and natural radionuclides. The analyses were carried out in cylindrical geometry, the mass of the samples was 180 g and the measurement time of a sample 4 h.

Of these, only the ^{137}Cs activity level was higher than the MDA (minimum detectable activity) threshold. ^{60}Co and ^{134}Cs could not be analysed due to low concentrations in relation to their significant decay. The reactor was shut down in 1997, about 10 years before the first soil investigation in 2006 and ^{60}Co concentration decreased by at least 4 times and ^{134}Cs by at least 32 times. Regarding their contribution during decommissioning, the soil could have been contaminated with gaseous and liquid effluents whose concentrations were initially comparable to those of ^{137}Cs , as reported by [1], but the meantime these were decayed. As for the ^{90}Sr , this is a beta pure emitting radionuclide difficult to detect especially in soil due to its high self-absorption. ^{90}Sr concentration measured in the effluents was lower than that ^{137}Cs , and it can be concluded that it has no influence on the supposed contamination of the soil [6].

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For natural radionuclides, the magnitude of exposure is determined by both ^{226}Ra progeny, which usually have the highest activity in the soil, and ^{232}Th progeny. The activity concentrations of ^{226}Ra and ^{232}Th were calculated based on the activities of the progeny (^{214}Bi and ^{214}Pb) and (^{208}Tl , ^{212}Bi , ^{212}Pb , ^{228}Ac), respectively. The measurements also revealed the presence of ^{40}K .

The results of the analysis are compared with the exclusion levels imposed by the basic domestic safety standards in force for the natural and the anthropogenic radionuclides (see Table 2) to demonstrate that the non-restrictive release criteria of the regulatory body's regime for land in the decommissioning area are met.

Table 2.

The exclusion levels for soil and sediments

No.	Radionuclide	Exclusion level* [Bq/kg]	Exclusion level** [Bq/kg]	Exclusion level*** [Bq/kg]
1	^{137}Cs	100	800	-
2	^{226}Ra	1000	200	200
3	^{232}Th	1000	200	200
4	^{40}K	10000	2000	-

Exclusion levels according with * [8], ** [9], *** [10]

4.1. ACTIVITY LEVELS OF THE ANTHROPOGENIC RADIONUCLIDES

^{137}Cs is the key fission product of ^{235}U and is an important anthropogenic radionuclide which has to be monitored in the area of a nuclear reactor. Thus, its concentration is systematically measured in all environmental samples, water, vegetation, soil, in IFIN-HH, by the Department of Life and Environmental Physics. Its special attention was due to reactor operation and decommissioning as well as the remaining contamination in Bucharest area due to the Chernobyl accident. The average values reported values for the IFIN-HH area were: (55.9±5.9) Bq/kg in 2008, (40.4±3.8) Bq/kg in 2009, (44.4±3.3) Bq/kg in 2010 [7]. A slight decreasing is observed, due to the decay ($T_{1/2} = 30.5$ y).

In our study, activity levels of ^{137}Cs in surface and depth soil were compared at the end of reactor operation and decommissioning to determine potential soil contamination in the reactor area. Thus, in 2020 at the end of reactor decommissioning, the highest concentration of ^{137}Cs in the surface soil (see Figure 4) was recorded in P4/8S (61.4 Bq/kg) and P4/7S (55 Bq/kg) points, near the sewerage for radioactive liquids. But the values were at about 10 times lower than in

2006 when the measurements showed that the soil is polluted due to reactor operation [2].

Meantime the soil was replaced with a clean one and ^{137}Cs concentration decreased. The mean value in 2020 was about 20.5 Bq/kg and the exclusion level of 100 Bq/kg stipulated in [8] was not exceeded. It is worth noticing that these values are comparable with those reported for IFIN-HH area.

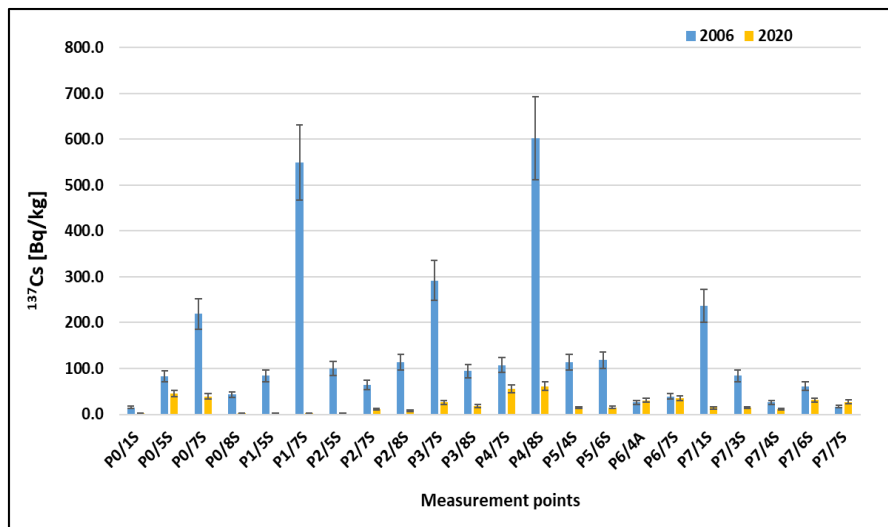


Fig. 4. ^{137}Cs activity concentration in the soil surface

Regarding ^{137}Cs activity in depth soil, the concentrations at the end of reactor operation in 2006 were higher than in 2020 for most samples, and the average value was 54 Bq/kg (see Figure 5).

In 2006, concentrations higher than 200 Bq/kg were found along the route of the radioactive effluents pipeline and maximum of 345 Bq/kg was recorded in P0/8A point [2].

However, according to the Fundamental radiological safety norms provision's [9] the exclusion level of 800 Bq/kg was not exceeded. At the end of reactor decommissioning in 2020, ^{137}Cs concentrations ranged between 2 - 59 Bq/kg (the uncertainty was 15%).

In the points (P4/7A, P5/6A, P6/4A, P7/4A) the activity concentrations were higher than in 2006, but the exclusion level of 100 Bq/kg [8] was not exceeded.

Soil activity in reactor decommissioning area

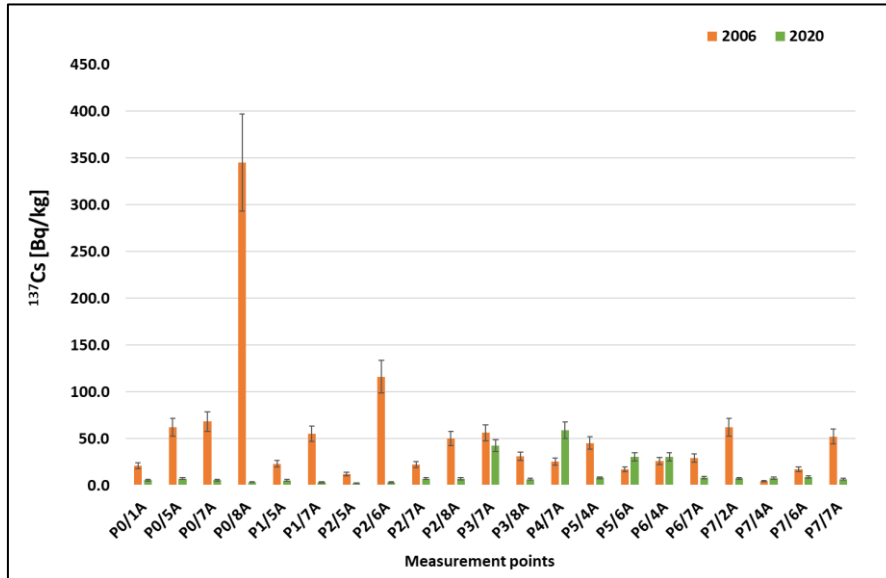


Fig. 5. ^{137}Cs activity concentration in depth soil

4.2. ACTIVITY LEVELS OF NATURAL RADIONUCLIDES

4.2.1 Activity levels of natural radionuclides in the surface soil

Gamma spectrometric analysis for the soil sample sampled from the surface soil performed both at the end of reactor operation [2] and decommissioning, revealed the presence of ^{226}Ra progeny (^{214}Pb and ^{214}Bi) and ^{232}Th progeny (^{208}Tl , ^{212}Bi , ^{212}Pb , ^{228}Ac) as well as ^{40}K .

In 2020, ^{226}Ra activity concentration in the surface soil ranged between 34.3 ÷ 45.4 Bq/kg and was evenly distributed around the mean value of 39 Bq/kg (see the Figure 6). Also, the mean concentration and was comparable with that reported for the soil from Romania (32 Bq/kg) in [11].

Concentration up to 2.7 times higher than in 2006 were found it due to the earth excavations in the decommissioning area. Maximum from 2020 (45.4 Bq/kg) recorded in P2/7S point near the radioactive liquid effluent sewerage was lower than those from 2006 (143 Bq/kg), recorded in the P6/7S point near non-operational Zero Power Reactor owned by the Bucharest Polytechnical University.

The exclusion levels stipulated by [8] (1000 Bq/kg) and [9] (200 Bq/kg) were not exceeded for both planned exposure situations (reactor decommissioning and operation).

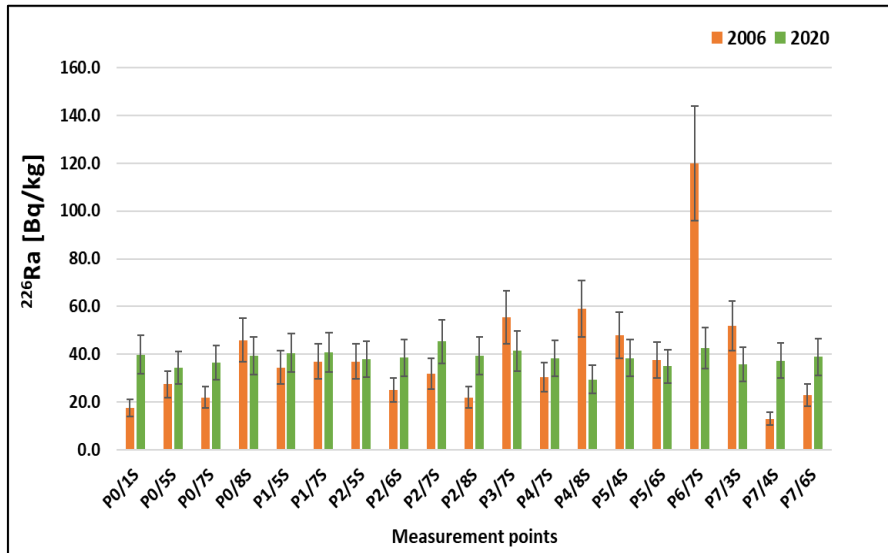


Fig. 6. ^{226}Ra activity concentration in soil surface

Regarding the ^{232}Th concentrations in soil surface a similar behaviour can be observed (see Figure 7).

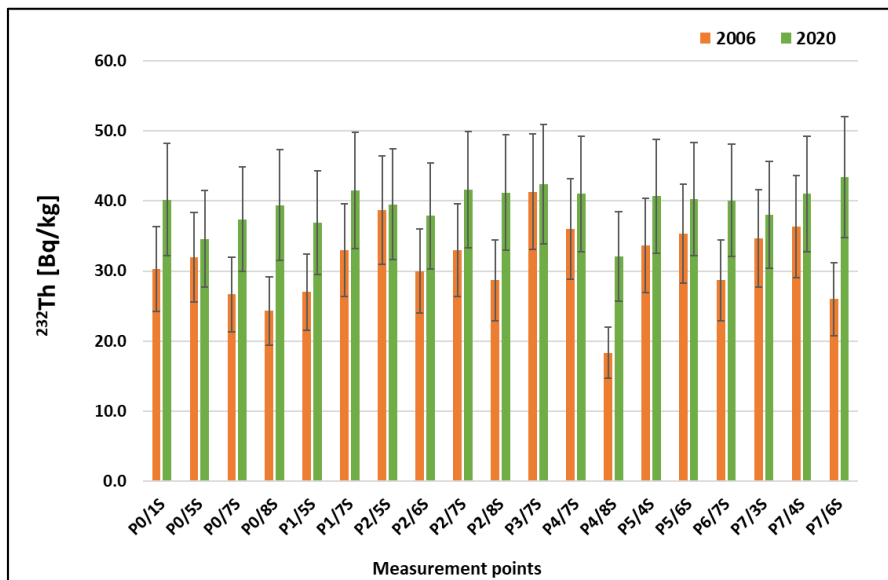


Fig. 7. ^{232}Th activity concentration in the soil surface

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In 2020, ^{232}Th average concentration was 39.4 Bq/kg and comparable with that for soil Romanian (38 Bq/kg) [11]. The maximum of 43.4 Bq/kg, was recorded near the Zero-Power Reactor (in P7/6S) and comparable with maximum from 2006 (41.3 Bq) recorded near the radioactive liquid effluent sewerage (in P3/7S). ^{232}Th concentrations from 2020 are up to 2.3 times higher than in 2006 due to soil excavation, but the exemption level of 1000 Bq/kg [8] was not exceeded.

Regarding ^{40}K , an average concentration of 531 ± 80 Bq/kg was recorded in 2020 and 492 ± 74 Bq/kg in 2006 in the surface soil from reactor decommissioning area. In the deep soil, the average in 2020 was of 511 ± 77 Bq/kg and 538 ± 81 Bq/kg in 2006.

The values were lower than the mean value of the years 2008, 2009 and 2010 for the IFIN-HH area reported in [8] and comparable with the ^{40}K mean concentration in soil for Romania of 490 Bq/kg [11].

4.2.2 Activity levels of the natural radionuclides in the deep soil

In 2020, ^{226}Ra concentration in deep soil (30 cm) (see Figure 8) ranged between $23 \div 40.5$ Bq/kg, and the mean was comparable to that reported for Romania in [11]. The peak was recorded near the sewer with radioactive liquid effluent. The exclusion level has not been exceeded [8].

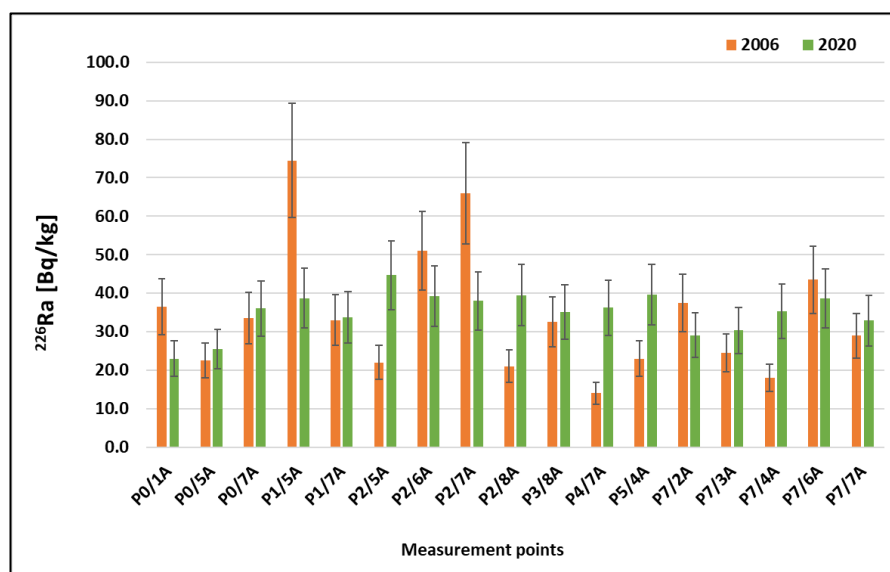


Fig. 8: ^{226}Ra activity concentration in deep soil

The activity concentration of ^{232}Th in the deep soil in 2020 (see Figure 9) ranged from 23.4 to 44.3 Bq/kg and the mean of 36.7 Bq/kg was comparable to the

value of 38 Bq/kg reported for Romanian soil in [11]. The peak was also recorded in the vicinity of the sewer with radioactive liquid effluent, but the exclusion level has not been exceeded [8].

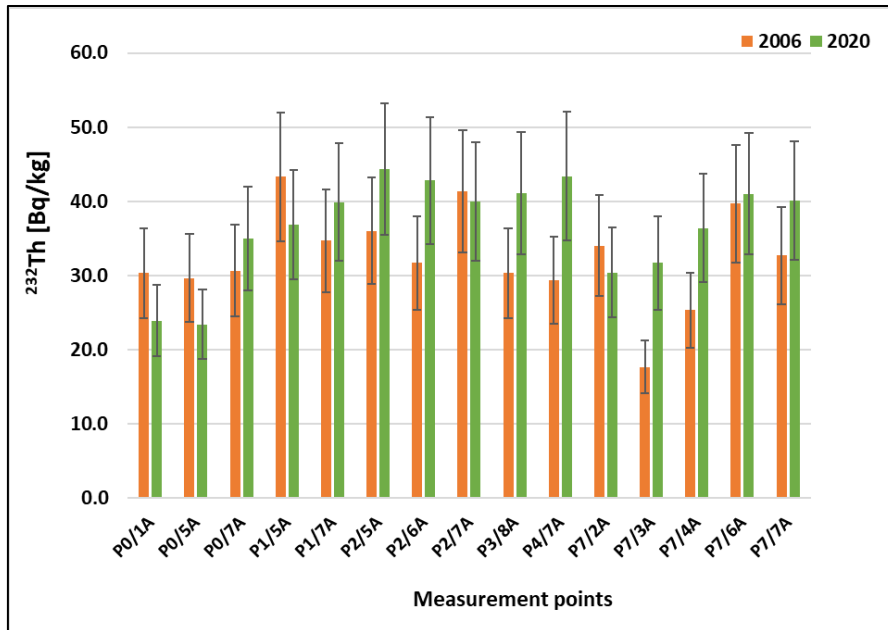


Fig. 9. ^{232}Th activity concentration in depth soil

4.2.3 Comparative analysis of activity levels for anthropogenic and natural radionuclides

The comparative analysis of the activity levels for the natural and anthropogenic radionuclides found in the soil samples at the end of the reactor operation and decommissioning was carried out to identify the background level for each radionuclide, distribution of activity levels due to sources of pollution, the relationship of "homogeneity or heterogeneity" between different pairs of radionuclides, as well as the "belonging" of the radionuclide at a process or a transfer path from source to the target.

The analysis was performed for the measuring points that have identical positions in the measurement plans (see fig 2 and fig 3) in the analysed periods (2006 and 2020).

The correlation graph between the natural and anthropogenic radionuclides $\Sigma^{226}\text{Ra} + ^{232}\text{Th} - ^{137}\text{Cs}$ are expressed in double logarithmic coordinates.

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In 2020, at the end of reactor decommissioning, from the correlation graph between the natural and anthropogenic radionuclides ($\Sigma^{226}\text{Ra}+^{232}\text{Th} - ^{137}\text{Cs}$, Figure 10), it can be noticed that the activity levels for both types of radionuclides are heterogeneously distributed (there were not recorded concentration higher than 100 Bq/kg).

For and average content of 76 Bq/kg for $\Sigma^{226}\text{Ra}+^{232}\text{Th}$, ^{137}Cs was bimodal distributed. In the group A, concentrations ranged between 1.3 ÷ 8.8 Bq/kg, and the mean was 5.1 Bq/kg; in the group B, the values were between 11.5 ÷ 61.4 Bq/kg and the mean value was 30.5 Bq/kg.

The average concentration of ^{226}Ra at the soil surface and depth are comparable and corresponds to the equilibrium with 3.3 ppm of natural uranium, the characteristic value of the natural background for soil [2]. Also, the ^{232}Th , concentrations are comparable and corresponds to the equilibrium with 8.75 ppm of natural thorium, the characteristic value of the soil background [2].

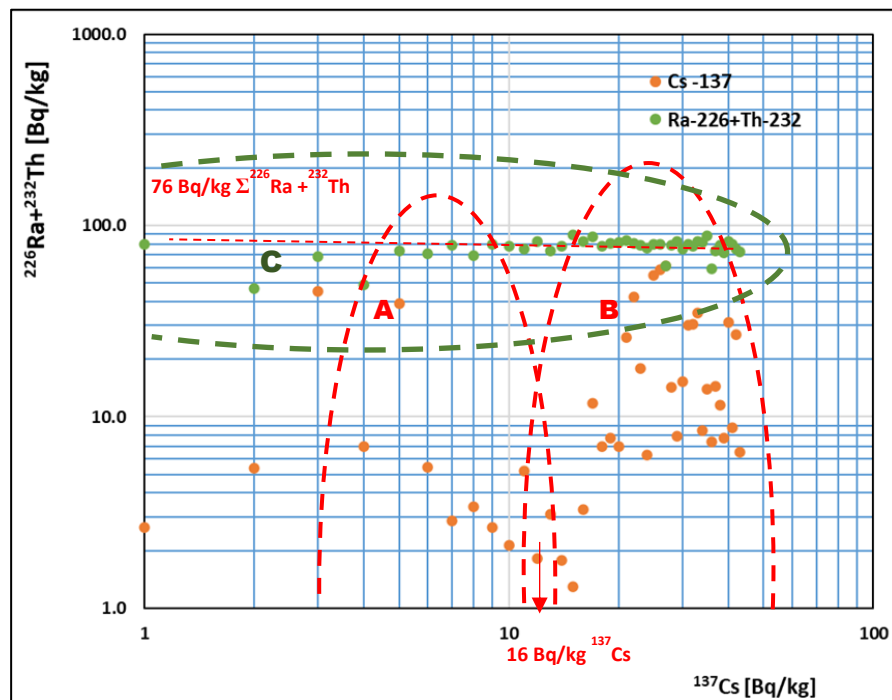


Fig. 10: $\Sigma^{226}\text{Ra}+^{232}\text{Th} - ^{137}\text{Cs}$ correlation graph in 2020

In 2006 at the end of the reactor operation, from the correlation graph between the the natural and anthropogenic radionuclides (see Figure 11) it can be noticed that

the activity concentration of the natural radionuclides expressed as a sum ^{226}Ra and ^{232}Th concentration is heterogeneously distributed in comparison with ^{137}Cs .

^{137}Cs concentration ranged between $4.4 \div 600.2$ Bq/kg for an average concentration of 69 Bq/kg for $\Sigma^{226}\text{Ra}+^{232}\text{Th}$. ^{137}Cs concentration was trimodal distributed. In the group A, the values range between $4.4 \div 31.0$ Bq/kg, with a mean of 20.4 Bq/kg. In the group B, values were between $39 \div 118$ Bq/kg with a mean of 76 Bq/kg. In the group C, concentrations were higher than 200 Bq/kg and the maximum was 602.0 Bq/kg. ^{137}Cs concentrations higher than the background (the "random" distributed values from group C from the Figure 11) were recorded due to radioactive liquids possible leakages from the pipelines who connected reactor with the intermediate storage tanks from the Radioactive Waste Treatment Plant.

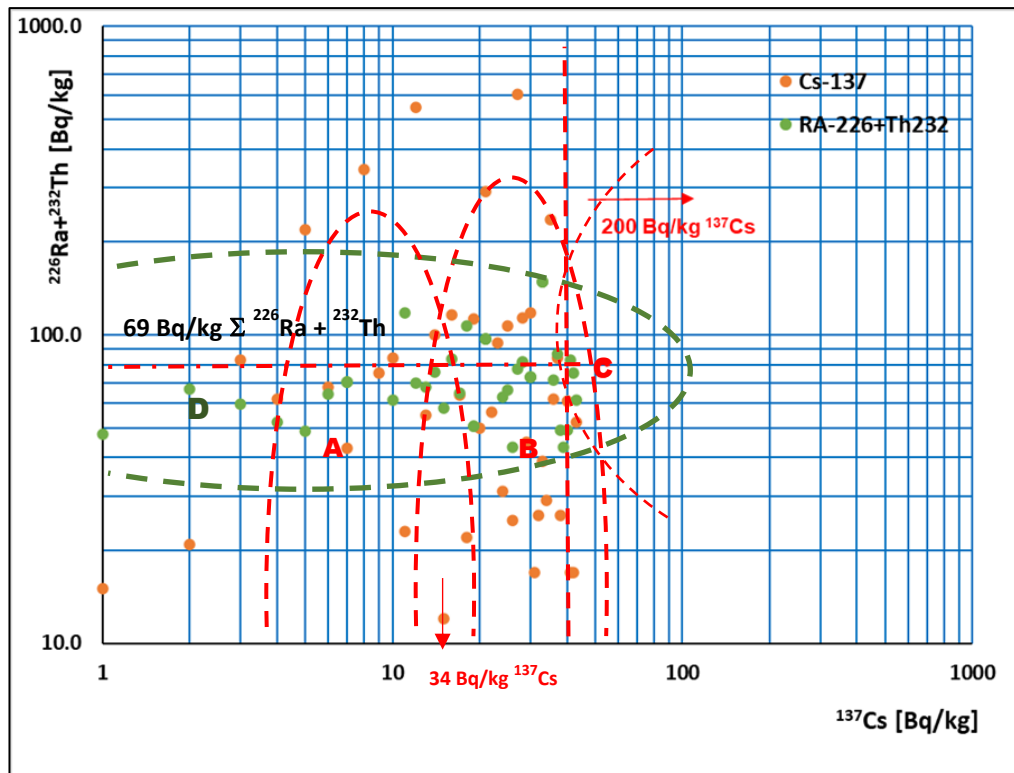


Fig. 11: $\Sigma^{226}\text{Ra}+^{232}\text{Th}$ - ^{137}Cs correlation graph in 2006 [2]

5. CONCLUSIONS

The soil activity levels of the natural and artificial radionuclides from the IFIN-HH VVR-S Nuclear Research Reactor area, were analysed both at the end of reactor operation (2006) and decommissioning (2020) in order to detect a potential soil pollution. The activity levels were obtained by gamma ray spectrometric measurements of samples taken both from the soil surface (10 cm) and depth (30 cm).

Regarding the activity levels of natural radionuclides ^{226}Ra and ^{232}Th , obtained by summing of the decay products activities, it can be observed that not exceed the exclusion levels provided by the Romanian legislation in force, 1000 Bq/kg in 2020 [8] and 200 Bq/kg in 2006 [9]. Also, the average concentration in soil is similar to that reported in [11] for Romania, 32 Bq/kg (^{226}Ra) and 38 Bq/kg (^{232}Th), respectively.

At the end of reactor decommissioning, the average concentration of ^{40}K in soil is comparable with the value of 490 Bq/kg reported in [11] for Romania.

At the end of reactor operation ^{137}Cs concentration in the soil from the reactor area was lower than the exclusion level of 800 Bq/kg [9], although areas with higher activities were recorded due to the experiments carried out during this period. Also, in 2020, at the end of reactor decommissioning, the ^{137}Cs exclusion level of 100 Bq/kg [8] was not exceeded.

Moreover, the ^{137}Cs low activity levels from 2020 emphasise that the decommissioning activities of the VVR-S Nuclear Research Reactor were properly performed and did not cause the soil contamination.

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