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PRELIMINARY CHARACTERIZATION OF IFIN-HH MAGURELE NUCLEAR SITE

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Magurele site includes nuclear facilities significantly influencing the environment, near a city, Bucharest, with 2 millions inhabitants: VVR-S research reactor, spent fuel wet storage, U-120 cyclotron, radioisotopical production center, radioactive west treatment plant, Helen subcritical assembly, etc. IFIN-HH as license holder of these installations, accordingly with the country legislation in force, must provide radiological surveillance results of the site and its influence area for sanitary police and for authorities in charge for environ-mental protection. The measurements are performed by γ spectrometry on the soil and vegetation samples in the influence area of the institute. The works are performed under IAEA Research contract 8735/RO, in the frame of Coordinated Research Program: "Characterization of environmental radioactivity in the influence area of IPNE Bucharest Magurele".

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1. INTRODUCTION

Different nuclear activities were developed on the Magurele site beginning fifties. The start-up of the research reactor type VVR-S in 1957 increased the radioactive pollution potential.[1]

IFIN-HH operates in the same site other nuclear facilities: cyclotron, TANDEM accelerator, subcritical assembly, radioisotope production center, spent fuel storage, radioactive waste treatment plant, nuclear medicine lab. In the last 40 years a large amount of radioactive materials was produced and manipulated.

On the Magurele site are also stored drums and packages containing historical radioactive wastes. There are also conditioned old drums, in advance degree of corrosion in the temporary storage. These were conditioned before the beginning of national repository operation.

Taking into account the future projects of the IFIN-HH concerning:

- decommissioning of VVR-S research reactor;
- refurbishment of the obsolete Radioactive Waste Treatment Plant (RWTP);
- dry storage of the VVR-S spent fuel;
- irradiator commissioning.

it is very important to clarify all the aspect of radioactive characterization of the site. The waste routes used by these activities must be strongly surveyed.

2. SURVEILLANCE

In IFIN-HH there is a specialized lab for environmental surveillance of the site and surroundings. A environmental monitoring system was developed [2] accordingly with new legislation of the country (Laws 134/1995 and 111/1996 regarding environmental protection and safe deployment of nuclear activities). Nuclear installations operating on the site release following maximum estimated radioactivity:

- in air:
  - VVR-S reactor 1000 Ci/annum
  - Radioisotope production center (venting) 100 Ci/annum
  - Nuclear medicine lab and RWTP 1 Ci/annum
  - Other facilities < 0.1 Ci/annum
- liquid effluents: RWTP treats all the liquid radioactive wastes. The amount of the effluents released in Ciorogirla river is about 1500 m^3 yearly, with the maximum permitted activity of 10^{-3} Ci/m^3.

The radioactive wastes (other than those from fuel cycle) are managed for all the country by IFIN-HH through RWTP Magurele and National Repository Baita-Bihor. The annual capacity of the treatment plant is around 1500 m^3 for low level aqueous wastes, 100 m^3 for low level solid wastes and 100 shield drums for medium level wastes. The National Repository has a capacity of about 20,000 standard drums, being placed in an old uranium mine.

The monitoring places and the sampling are established by internal documents. These points cover a large area, taking into account the meteorological factors and nuclear activities. The description of the prelevation points, the sample types and the sector are presented in Table 1.

Surveillance frequency for all type of samples depends by the characteristics of the average time of residence and life time in a exposure route, for each nuclide. The used methods for characterization of the radioactivity of the influence area of IFIN-HH are β global and γ measurements.

A special attention is awarded to the area Reactor&RWTP, including away-far-reactor (AFR) storage of spent fuel.

3. RADIOLOGICAL CHARACTERIZATION

Presently, the institute applied an environmental concept.[2] There are established 19 points of monitoring for sampling of vegetation, soil and waters. This document also provides the survey frequency, the methods and the detection limits.

Taking into account the future development of the nuclear activities in the IFIN-HH zone concerning VVR-S research reactor decommissioning, modernizing of RWTP, the site decontamination of old fortresses, there is absolutely necessary an improvement of this environmental surveillance system. By the above mentioned activities will be manipulated a large amount of liquid and solid radioactive waste and also will appeared aerosols, being potential radioactive pollutants of the IFIN-HH influence area. The necessary radiological site characterization must be more detailed by the increasing of the surveyed points and the using of new measuring techniques.

In this respect, in 1996, we established nine new points of surveillance, gathered in Table 2. In 1997 we considered four additional points in the RWTP yard. In Figure 1 are presented all the points for prelevation.

The detailed research objectives are:

- characterization of radioactive contamination in the IFIN-HH influence area in air, soil, vegetation, liquid effluence;
- determination of radioactive migration in environment;
- determination of soil-plant transfer factor in field conditions for ^{137}Cs and ^{60}Co;
- modeling of radionuclide migration.

We defined the concept strategy of site characterization by improving of the environmental surveillance. This improvement is based on the identification of the contaminated area boundaries, in the following zones:

- VVR-S research reactor
- RWTP
- old military fortresses

using portable instrumentation.

TABLE 1
Monitoring points and sampling

Point	*Description of place*	*Sample*	*Sector*
A	1st channel of Reactor	- water - sediment - vegetation - soil	S
B	2nd channel IFA	- water	S
C	Ciorogirla river - upstream	- surface water - sediment	SSW
D	Ciorogirla river - downstream	- surface water	ESE
E	Used domestic water plant	- water - sediment	WSW
F	ICAB plant	- water - sediment	S
G	1st group IFIN-HH	- drinkable water	WSW
H	2nd group IFA	- drinkable water	
I	Village fountains	- water piping in 1st channel - Reactor (ground water) - water piping in 2nd channel - IFA (ground water)	ESE ESE
J	Surveillance bore holes	ground water in the Reactor area	
K	IFA park	- soil - vegetation - sediment	WSW
L	Helen subcritical assembly	- soil - vegetation	WSW
	Bucharest-Titan (background)	- soil - vegetation	ENE
M	Forest	- soil - vegetation	WSW
	Village (channels area)	- soil - vegetables - cereals - fruits - milk	S
N	HV supply	- soil - vegetation	WNW
O	Pumping station	- soil - vegetation	NW
P	Mechanical workshop	- soil - vegetation	N
R	Lab. No. 8	- air	NW

TABLE 2
Vegetation and soil samples characteristics

Point	*Sample identification*	*Sample mass (g)*	*Prelevation point*
P1	veg1 soil1	61.18 216.13	Nuclear Medicine Lab
P2	veg2 soil2	54.75 215.52	Cyclotron building (left side)
P3	veg3 soil3	60.85 196.50	Reactor plateau (center)
P4	veg4 soil4	56.25 206.40	Reactor auto-gate (left)
P5	veg5 soil5	52.55 195.11	Reactor auto-gate (right)
P6	veg6 soil6	56.89 195.00	Spent fuel storage (left)
P7	veg7 soil7	60.99 193.05	Spent fuel storage (right)
P8	veg8 soil8	61.00 203.35	Reactor - RWTP route (5 m right from plant gate)
P9	veg9 soil9	60.88 193.62	Reactor - RWTP route (5 m left from plant gate)
P10	veg10 soil10	89.09 233.00	Waste pond (underground)
P11	veg11 soil11	110.28 236.50	Surface waste storage
P12	veg12 soil12	100.6 219.22	300 m^3 ponds
P13	veg13 soil13	62.41 198.60	Heating plant

The contaminated areas are marked for sample prelevation. The radiological characterization of these areas is performed by "in-laboratory" α, β and γ measurements. The absolute methods for determination of radioactive concentrations in inventory are used.

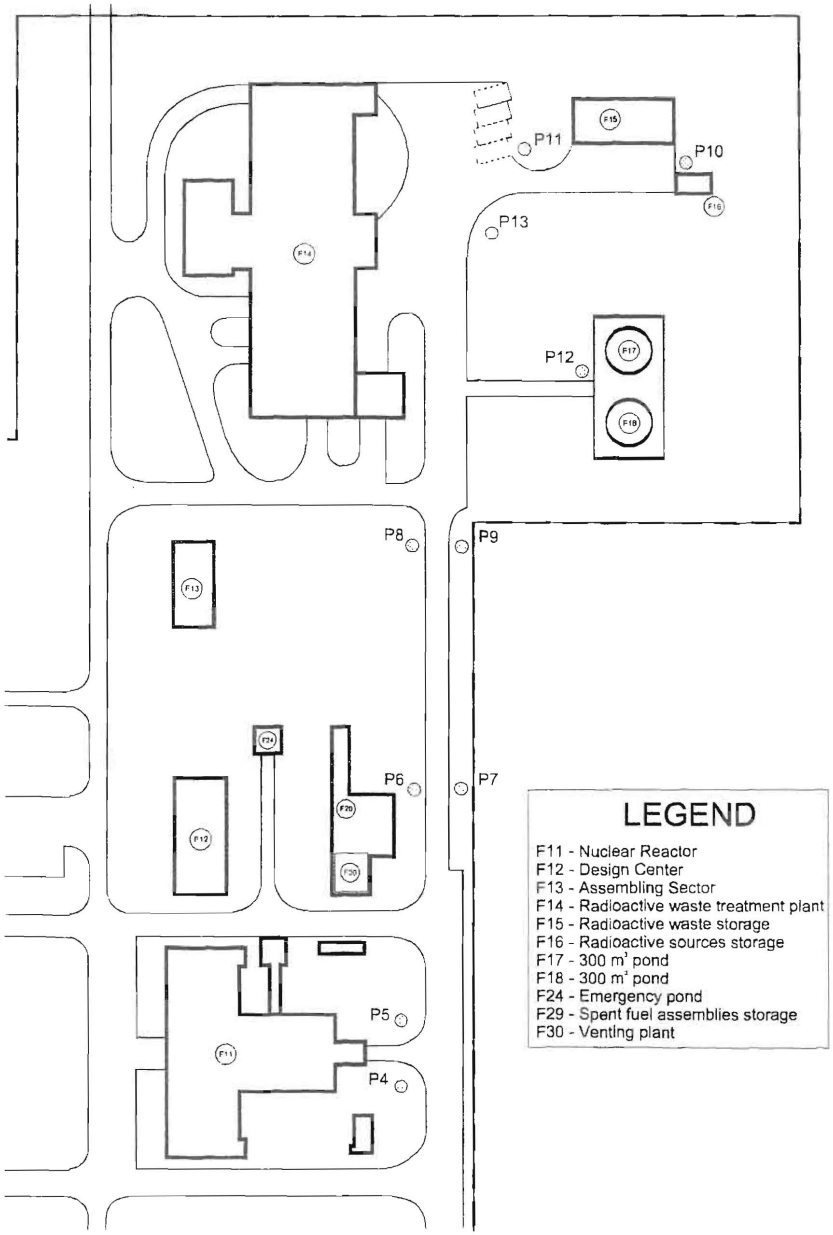

Figure 1

The high resolution gamma spectrometer NOMAD PLUS used in this research contract was delivered by generous assistance of International Atomic Energy Agency, in the frame of Technical Assistance Project ROM/9/017.

For the portable system SMART-200 is provided with the detectors for α contamination, β-γ contamination and γ exposure (ranges: 20 - 70 keV, 40 keV - 1 MeV). This system was also obtained by the Agency assistance.

4. RESULTS

For the preparation of standard as well as for solid material measuring was chosen a plastic box, with 72.7 mm diameter and 36.5 mm height. These boxes are currently available on the internal market, serving for medicine packing. The box thickness vary about 0.1 mm, their weight being about 19g. The weight variation is about 2%, due the nonuniformity of fabrication.

The materials used for preparation of standards are:

- leaves: gathered from reactor yard, dried at 90 Celsius degrees and went through a mill;
- soil: taken for 1m deep, in village Mosteni-Teleorman; dried at 90 Celsius degrees and broken at a very fine granulation;
- water: distillate.

These materials are standard supports in order to avoid the density differences between standard and environmental sample. The standard solutions of ^{152}Eu, ^{241}Am, ^{137}Cs have been mixed with above mentioned supports in order to be homogenous materials. The radioactive solutions are certified by Radiation Metrology Laboratory, with $\pm$ 5% precision. These is used a Mettler balance for weighting of radioactive solutions.

The soil and vegetation samples are processed by a similar procedure as standards. These samples are sealed in plastics box. The sample densities are comparable with those of standards in order to avoid the connection for variation of efficiency versus density.

The measurements have been performed in plan-contact by high resolution gamma-spectrometer NOMAD PLUS its relative efficiency being 42.5%. The spectrometer is placed in lead shielding with dimensions 1m x 1m x 1m. The gamma background has been reduced by about ten times.

For the determination of gamma-spectrometer calibration factors used for in-situ measuring was chosen a circular geometry (1m radius). The ring was divided in 24 equal sectors. The used reference sources are the following:

^{152}Eu with activity 370.0 kBq
^{60}Co with activity 306.9 kBq
^{137}Cs with activity 255.7 kBq
^{241}Am with activity 338.1 kBq
(on the 1st of July 1985)

and

^{51}Cr with activity 261.9 kBq
(on the 25th of October 1996).

The calibration was performed for liquids, gels, soil and vegetation. A special device was designed for the setting up in different positions for various source-crystal distances. There was also designed a special recipient for liquids.
The calibration data for NOMAD spectrometer in terms of efficiency are given in references [3] and [4].

In the following table are given the standards for soil and vegetation.

Reference source	Density (g/cm^3)	Activity (Bq)*
eusol1	1.45	733.40
euveg1	0.61	584.41
euveg2	0.40	542.14

* measured at 01.08.1996 with an error of ±5%

The results for soil and vegetation measuring in IFIN-HH influence area are obtained by processing of γ spectra. By using of GAMMA VISION code ("Analyze" option) have been identified: peak center, FWHM, FWTM and also net area. The radioisotope was identified using the energy calibration, by means of the nuclear library data provided by above mentioned code. The content (in Bq) for the radioisotopes identified in samples was determined by means of the efficiency calibration.

The precision of the identification of the isotope energies is ±0.5keV, corresponding to one channel deviation (for a spectrum recorded in the 4096 channels mode). This uncertainty of measuring creates some difficulties in radioisotope identification, using the automatic processing. There was applied a semi-automatic procedure processing, in order to avoid this miss-determination.

Applying the above mentioned procedure has been reached the following radioisotopes: ^{40}K, ^{7}Be, ^{137}Cs, ^{134}Cs, ^{212}Pb, ^{214}Pb, ^{131}I and ^{214}Bi. The vegetation samples radioactivity (in Bq/g) was given in Table 3. The radioisotopes due the vegetation contamination as ^{137}Cs, ^{134}Cs, ^{131}I have been identified. The identified Bi and Pb isotopes are from soil composition. They passed through vegetation. The presence of ^{131}I in vegetation shows a small contamination with this radioisotope in some points (2, 3, 4, and 8).

Table 4 gives the radioactivity values (in Bq/kg) for soil samples. The fission products ^{137}Cs, ^{134}Cs has been identified. The soil samples no.6 and 7 taken near the spent fuel storage have values two times more than all samples in IFIN-HH zone for ^{137}Cs.

A field calibration was performed by a combined method (experimental and theoretical) recommended by I.K.Helfer & K.M.Miller in reference [5].

The basic calibration equation for "in-situ" spectrometry can be expressed in terms of peak count rate (N), activity or inventory in the soil (A) and uncollided flux (Φ) as:

$$N_f/A = (N_f/N_0)(N_0/\Phi)(\Phi/A) \qquad (1)$$

where the fundamental calibration parameters are expressed in ratios as follows:

- N_f/A is the total-absorption peak count rate (cpm) in the spectrum at the energy of a particular nuclide γ transition per unit inventory ($Bq\cdot m^{-2}$) or concentration ($Bq\cdot g^{-1}$) of that nuclide in the soil,
- N_f/N_0 is the angular correction factor of the detector at that energy for a given source distribution in the soil,
- N_0/Φ is the peak count rate (cpm) per unit uncollided flux ($\gamma\cdot cm^{-2}\cdot s^{-1}$) for a parallel beam of γ rays of the same energy that is incident-normal to the detector face, and
- Φ/A is the total uncollided flux ($\gamma\cdot cm^{-2}\cdot s^{-1}$) at that energy arriving at the detector per unit inventory or concentration of the nuclide in the soil.

In order to obtain the calibration factor (N_f/A) for a particular nuclide, the three quantities (N_f/N_0, N_0/Φ and Φ/A) are determined separately. The first two terms are detector dependent and can be determined experimentally, while the latter can be calculated on a purely theoretical basis.

The value of N_0/Φ at a particular energy is obtained by counting a γ-emitting point source of known strength, placed at a distance of at 1m from the detector face to simulate a parallel beam of normally incident radiation. The work is in progress.

The value of N_f/N_0 for a particular energy and source distribution is calculated from:

$$N_f / N_0 = \frac{\int R(\theta)\Phi(\theta)d\theta}{\int \Phi(\theta)d\theta}, \qquad (2)$$

where $R(\theta)$ is the peak count rate for γ rays of energy E at angle θ, relative to the peak count rate at $\theta=0^0$ (normal incidence) and $\Phi(\theta)$ is the γ-ray flux at energy E at angle θ.

The coefficients Φ/A and N_0/N_f are presented in reference [4]. The results for various distributions in soil are also given in the same reference, based on the calibration factors.

TABLE 3. Specific activity (Bq/kg) for each isotope present in vegetation samples

	Isotope	96veg1	96veg2	96veg3	96veg4	96veg5	96veg6	96veg7	96veg8	96veg9
Sample mass (g)		61,18	54,75	60,85	56,25	52,55	56,89	60,99	61,00	60,88
	Be7	227,88	446,08	193,96	202,66	400,54	136,26	127,60	130,52	258,52
	K40	310,80	241,54	805,10	599,06	137,58	802,78	1152,30	905,86	320,68
	Cs134	26,18	40,58	22,34	25,76	24,00	15,38	13,68	14,76	13,22
	Cs137	6,04	10,86	3,96	4,56	3,60	7,32	13,12	6,46	7,44
	Pb212	0,00	27,38	0,00	3,62	10,18	0,00	0,00	0,00	0,98
	Pb214	28,40	70,46	19,52	16,44	57,50	45,14	32,70	42,04	52,92
	I131	5,64	6,20	4,52	7,60	0,00	0,00	0,00	6,50	3,86
	Bi214	1,18	1,02	0,00	0,00	0,98	0,84	0,88	1,20	1,28
	Ac228	1,18	12,98	59,50	1,32	6,88	0,00	0,00	0,00	2,06
	TOTAL	**607,30**	**857,10**	**1108,90**	**861,02**	**641,26**	**1007,72**	**1340,28**	**1107,34**	**660,96**

TABLE 4. Specific activity (Bq/kg) for each isotope present in soil samples

	Isotope	96sol1	96sol2	96sol3	96sol4	96sol5	96sol6	96sol7	96sol8	96sol9
Sample mass (g)		216,13	215,52	196,50	206,40	195,11	195,00	193,05	203,35	193,62
	Ra226	167,08	149,74	210,72	186,76	213,98	195,74	166,66	198,44	185,68
	Pb212	54,16	57,90	67,92	63,78	74,06	60,04	40,17	63,48	56,22
	U235	122,80	141,62	152,16	111,92	194,46	167,58	88,38	143,10	161,46
	Tl208	18,88	17,72	19,64	18,02	20,92	18,88	10,46	20,46	17,04
	Pb214	53,52	50,40	57,74	59,50	60,54	51,38	51,60	60,36	48,30
	Bi214	40,02	34,40	39,46	40,34	43,02	35,60	33,14	43,80	30,00
	Cs137	200,10	150,36	195,04	244,60	169,88	493,36	322,54	214,84	182,96
	Bi212	33,84	29,00	40,54	31,54	36,26	29,56	18,54	38,62	33,66
	Cs134	7,22	6,04	6,72	7,94	6,76	12,54	9,74	7,86	6,00
	Ac228	52,46	41,92	50,06	48,14	52,60	44,28	25,34	48,78	40,60
	K40	830,78	822,32	930,60	916,98	903,38	862,44	756,56	914,16	815,04
	TOTAL	**1580,86**	**1501,42**	**1770,60**	**1729,52**	**1775,86**	**1971,40**	**1523,13**	**1753,90**	**1576,96**

5. CONCLUSIONS

1. The environmental surveillance strategy in IFIN-HH influence area must be defined and updated versus the nuclear activities that will be developed on the site accordingly with international practices [6], [7], [8], [9].
2. This paper work presents the methods developed by means of the instrumentation get by Agency's assistance for:
   - γ spectrometry
   - measuring of α, β and γ contamination (with portable equipment), this instrumentation being calibrated.
3. The measured soil and vegetation samples have been prelevated in currently surveyed zones and that including radioactive waste routes and temporary storage for spent fuel.

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