STUDY OF ENVIRONMENTAL RADIOACTIVITY ALONG THE ROMANIAN DANUBE BANK

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Abstract: *A study of environmental radioactivity along the Romanian Danube bank was performed by Radioprotection and Environmental Protection Laboratory from Institute for Nuclear Research – Pitesti, between August 2006 and September 2007, within the framework of the government funded research project: "Research network for integrated monitoring of the radioactivity and environment related isotopes throughout the Romanian Danube basin". The paper contains the most important conclusions and some correlations and interpretations of the obtained results. The measurements and samples collection were made according to a monitoring plan which included locations along the Danube River and its main tributaries or other locations of interest. The targeted radionuclides were ^{137}Cs, ^{40}K, ^{235}U and the gamma emitting radionuclides from the natural series of the ^{238}U and ^{232}Th. Results included in this study concern the dose rate measurements, the gamma emitting radionuclides concentrations measured by gamma-spectrometry in water, sediment and soil samples, the in-situ gamma-spectrometry and the gross beta and gamma concentrations measured in sediment samples.*

Keywords: *radioactivity monitoring, Romanian Danube basin.*

1. INTRODUCTION

Near the Romanian Danube basin operate several nuclear facilities, among which the most important are the Paks NPP (Hungary), Kozloduy NPP (Bulgaria) and Cernavoda NPP (Romania). These, but not only, may be potential sources of radioactive contamination of the environment. Knowledge of environmental radioactivity level is required both during normal operation of nuclear facilities, and in case of an unwanted nuclear accident. Establishment of the reference values of key parameters that characterize the environmental radioactivity level along the Romanian Danube bank was one of the aims of the research project "Research Network for integrated monitoring of the radioactivity and isotopes related environment throughout the Romanian Danube Basin". The radiological characterization was planned to be performed in three stages over a period of approximately one year, in locations distributed along the entire Danube's route through Romania, of approximately 850 km.

2. RADIOLOGICAL MONITORING PLAN

The radiological characterization of the Romanian Danube basin was made in three phases (August 2006, March 2007 and September 2007) and consisted in:

- dose rate measurements;
- in-situ gamma spectrometry measurements for determination of the gamma emitting radionuclides concentrations;
- sampling of soil, water and sediment and determination of the gamma emitting radionuclides concentrations by high resolution gamma ray spectrometry in laboratory;
- sampling of water and sediment and determination of the gross beta and gamma concentrations.

Two types of monitoring locations were considered (Fig. 1): the A - type locations, along the Danube river (Ieselnita, Gura Vaii, Bechet, Tr. Magurele, Calarasi, Seimeni mila 292, Giurgeni, Tulcea ponton) and the B - type locations, on Danube's main romanian tributaries (Toplet – Cerna river, Filiasi – Jiu river, Islaz – Olt river, Oltenita – Arges river, CNE Cernavoda – cooling water evacuation channel).

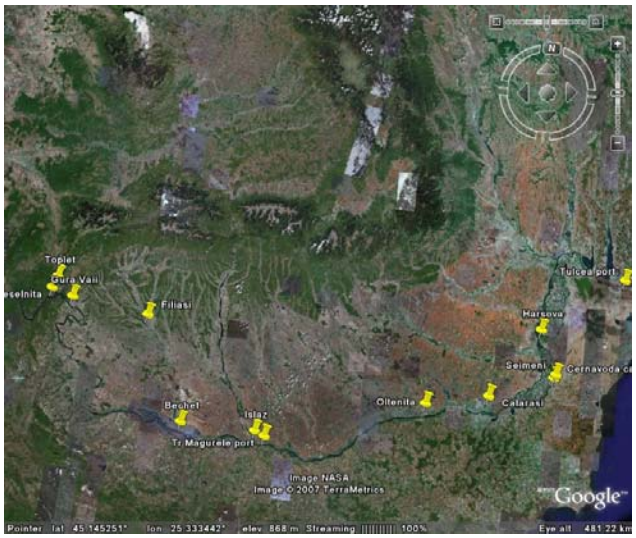

Fig. 1. Distribution of the monitoring locations

The targeted radionuclides were Cs-137, K-40, U-235 and the gamma emitting radionuclides from the natural series of the U-238 and Th-232.

3. INSTRUMENTS AND MEASUREMENT METHODS

Dose rate measurements were made 1 m above the soil with a portable spectrometric analyzer, InSpector type, model 1000 from Canberra Instruments Co., with a 2" NaI probe, calibrated in H*(10).

In-situ gamma spectrometry measurements were made with a portable spectrometric chain consisting of a coaxial HPGe detector 40% relative efficiency and an integrated gamma spectrometric analyzer, model digiDART from ORTEC. Energy range was 60 - 1840 keV. The calculation model was based on an exponential distribution characterized by a distribution parameter $\alpha/\rho = 0.3$ for Cs-137 (originating mostly from the Chernobyl accident fallout) and a homogenous distribution for the natural radionuclides.

For the gamma spectrometric measurements, the sediment and soil samples (taken from the same locations where in-situ gamma spectrometric measurements were made) were processed in laboratory by drying in open atmosphere, screening, grinding and drying in oven (110^{o}C) to constant weight. After a new grinding and screening with a fine sieve, required amounts of sample were putted in plastic boxes and measured. The measurements were performed with a spectrometric chain consisting of a coaxial HPGe detector 25% relative efficiency with low background shielding and integrated gamma spectrometric analyzer, InSpector type, model 1270 from Canberra Instruments Co. Energy range was 30 - 1700 keV. Acquisition time of the spectra was chosen as to achieve detection limits well below the exemption levels for many of the targeted radionuclides. If the detection limit exceeded measured value or the radionuclide was not found in a sample, one indicated the value of the limit of detection calculated with the Currie algorithm [1]. The radionuclides concentration in sediment and soil samples is reported against the mass of dry soil.

Gross beta measurements were performed with a low background alpha/beta radiometer, model MPC 9300 from Protean Instrument Corporation, efficiency calibrated with a set of KCl sources.

The uncertainties for all measurements were estimated only for the measurement phase and are indicated with a coverage factor k = 1.

4. RESULTS AND DISCUSSIONS

All results, for each of the three campaigns and for each of the thirteen monitoring locations, are widely presented in the publication [2]. In this paper registered results of the followed radiological parameters are generally synthesized as average values and variation intervals, calculated for each campaign, to support discussions and conclusions that will follow.

The results of the dose rate measurements are plotted in Figure 2. The values of the dose rate for A type locations are distributed between 30 and 63 nSv/h, with an average value of the 42 nSv/h and a standard deviation of the 7.5 nSv/h. The distribution interval for B type locations was between 30 and 60 nSv/h, with an average value of 46 nSv/h. The average values of the dose rate for each of the three campaigns of measurements in the A type locations were: 43 nSv/h for august 2006, 39 nSv/h for march 2007 and 44 nSv/h for september 2007.

No	Location
1	Ieselnita
2	Cerna Toplet
3	Gura Vaii
4	Filiasi
5	Bechet
6	Islaz
7	Tr. Magurele
8	Oltenita
9	Calarasi
10	Canal CNE
11	Seimeni mila 292
12	Giurgeni
13	Tulcea ponton

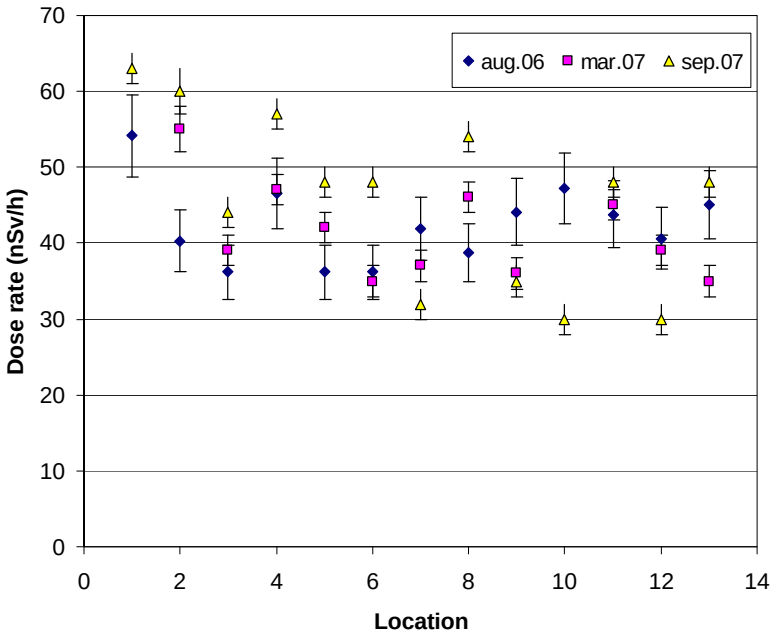

Fig. 2. The results of the dose rate monitoring

The values of gamma emitting radionuclides concentrations in sediment samples (determined by high resolution gamma ray spectrometry) and the values of the gamma emitting radionuclides concentrations in soil (determined by gamma in-situ measurements) are presented as averaged values on the all monitoring locations, for each campaign in Tables 1 and 2.

Table 1. The average values of gamma emitting radionuclides concentrations in sediment samples

Radionuclide	Average concentration (Bq/kg)		
	Aug. 2006	Mar. 2007	Sep. 2007
K-40	541 ± 117	501 ± 82	496 ± 94
Cs-137	6 ± 3	5 ± 3	4 ± 3
Tl-208	19 ± 6	28 ± 9	11 ± 3
Bi-212	35 ± 9	38 ± 8	40 ± 10
Pb-212	33 ± 10	33 ± 10	36 ± 11
Bi-214	26 ± 8	24 ± 7	25 ± 7
Pb-214	30 ± 8	26 ± 8	29 ± 8
Ra-226	58 ± 10	53 ± 19	25 ± 9
Ac-228	34 ± 10	34 ± 10	34 ± 10
U-235	4 ± 1		2 ± 1

Table 2. The average values of gamma emitting radionuclides concentrations in soil

Radionuclide	Average concentration (Bq/kg)	
	Mar. 2007	Sep. 2007
Be-7	683 ± 193	923 ± 214
K-40	466 ± 62	430 ± 38
Tl-208	23 ± 9	17 ± 4
Pb-212	40 ± 7	33 ± 11
Pb-214	46 ± 8	36 ± 11
Bi-212	50 ± 16	47 ± 15
Bi-214	42 ± 7	44 ± 13
Ra-224	56 ± 17	45 ± 11
Ra-226	128 ± 85	115 ± 86
Ac-228	41 ± 6	46 ± 11
Pa-234	17 ± 4	37 ± 17

Table 3 contains the results of the gross gamma and beta emitting radionuclides concentrations for sediment samples.

Both the measured values of the dose rate and gamma emitting radionuclides concentrations are within the limits of the natural background. From the measurement results of the gamma emitting radionuclides concentrations in soil and sediment samples in each location one can see that a good equilibrium between the radionuclides from the natural series of Th-232 (Tl-208, Bi-212, Pb-212, Ac-228) and U-238 (Pb-214, Bi-214, Th-234) is present. This equilibrium is illustrated in Table 4, where the correlation coefficients between the radionuclides concentrations are shown in sediment samples (from August 2006 campaign). A good correlation between the gamma dose rate and the K-40 concentration is also seen.

The correlation between the gamma dose rate and its main natural radioactivity contributors

This study was performed to identify the gamma radionuclides which have a major contribution to the dose rate. Nuclear data concerning gamma decay and the specific gamma emission for each identified radionuclide in the sediment samples were used (Table 5). For each radionuclide, the number of gamma photons per kg of sediment was calculated using the averaged value of the radionuclide concentration for all sediment samples (also, the data from

August 2006 campaign were used). As can be seen in Table 5, K-40 and Tl-208 are the main contributors to the global gamma emission.

Table 3. Gross gamma and beta emitting radionuclides concentrations for sediment samples

Location	Gross gamma concentration (Bq/kg)	Gross beta concentration (Bq/kg)	Discrepancy (%)	Gross gamma concentration (Bq/kg)	Gross beta concentration (Bq/kg)	Discrepancy (%)
	August 2006			September 2007		
Ieselnita	1003	858	14	1004	981	2
Cerna	838	882	-5	935	939	0
Filiasi	851	921	-8	769	849	-9
Bechet	570	544	5	896	909	-1
Islaz	529	520	2	1048	981	7
Tr. Magurele	666	620	7	542	529	2
Oltenita	912	840	8	616	638	-3
Calarasi	608	574	6	923	837	10
Canal CNE	798	791	1	739	704	5
Seimeni	832	779	6	728	710	3
Giurgeni	554	592	-7	655	770	-15
Tulcea	833	921	-11	882	577	53

Table 4. Correlation coefficients for radionuclides concentrations in sediment samples

	K-40	*Tl-208*	*Bi-212*	*Pb-212*	*Bi-214*	*Pb-214*	*Ac-228*	*Th-234*	*Cs-137*
K-40	1								
Tl-208	0.57	1							
Bi-212	0.72	**0.79**	1						
Pb-212	0.70	**0.96**	**0.91**	1					
Bi-214	0.57	0.94	0.85	0.95	1				
Pb-214	0.64	0.92	0.84	0.95	**0.98**	1			
Ac-228	0.71	**0.95**	**0.85**	**0.96**	0.96	0.95	1		
Th-234	0.54	0.82	0.83	0.85	**0.89**	**0.84**	0.83	1	
Cs-137	0.81	-0.06	0.18	0.07	-0.001	0.10	0.20	-0.03	1
H(10)*	**0.70**	0.37	0.56	0.55	0.32	0.43	0.39	0.35	0.32

In the assumption that the radioactivity content of the soil around the monitoring locations is similar to the one of the collected sediment (generally, the bank sediment has an erosion character), the dose rate should be correlated with the main gamma emitting radionuclides concentration. Fig. 3 shows the representation of the dose rate function of the K-40 content in the sediment samples and one can see that the correlation is acceptable.

Using the computation methodology described in *External Exposure to Radionuclides in Air, Water, and Soil*, Federal Guidance Report 12 of the U. S. Environmental Protection Agency [3] one can estimate the dose rate in the assumption of a quasi-infinite uniform distribution of the natural radionuclides in soil. The conversion factors recommended by Kocher [4] and FGR 12 are presented in Table 6.

Table 5. Nuclear data concerning gamma decay of the identified radionuclides in the sediment samples

Radionuclide	No. of γ photons / 100 disintegrations*	Energy (keV)	No. of γ photons/kg	keV/kg
K-40	11	1460.8	58	84636
Cs-137	90	661.7	5	3200
Tl-208	12	860.4	2	2072
	22	510.8	4	2129
	84	583.1	16	9496
	100	2614.7	19	50450
Bi-212	12	727.2	4	3039
Pb-212	11	74.8	4	266
	15	10.8	5	56
	18	77.1	6	462
	45	238.6	15	3542
Bi-214	15	1120.3	4	4398
	16	1764.5	4	7243
	46	609.3	12	7308
Pb-214	10	77.1	3	242
	14	10.8	4	44
	19	295.2	6	1705
	37	351.9	11	3928
Ac-228	11	338.3	4	1313
	17	969.1	6	5503
	28	911.1	9	8623
	39	13.0	13	174
U-235	11	143.8	0	60
	31	13.0	1	16
	54	185.7	2	401

*Only gamma energies with an emission probability better than 10% where included

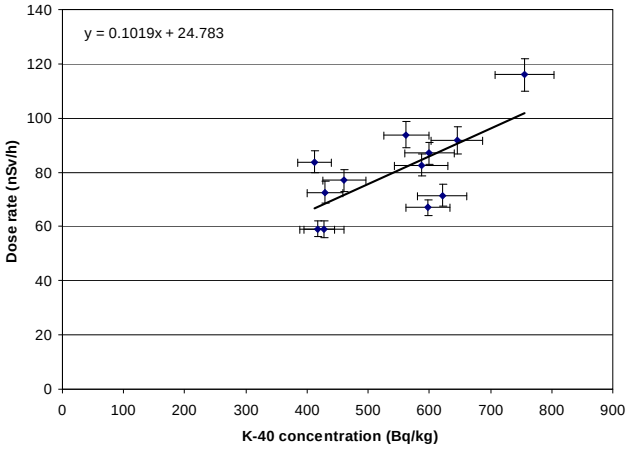

Fig. 3. Dose rate function of the K-40 content in the sediment samples

Table 6. Conversion factors for quasi-infinite uniform distribution in soil

Radionuclide	Conversion factor (nSv/h per Bq/kg)
K-40	0.058
Tl-208	1.264
Bi-212	0.064
Pb-212	0.037
Bi-214	0.539
Pb-214	0.072
Ac-228	0.327
Th-234	0.001

As can be seen in Fig. 4, the estimated dose rates are in good agreement with the dose rates measured in monitoring locations.

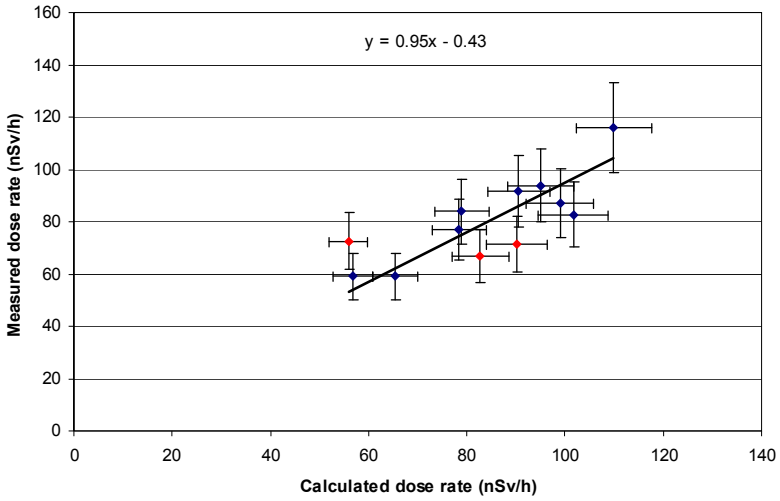

Fig. 4 Graphic representation of measured dose rates function of calculated dose rates

This result may be interpreted as an argument for the representativeness of the sediment samples, proving the uniformity of the gamma emitting natural radionuclides distribution in the vicinity of the sampling locations.

Comparative analyses of the gamma in-situ measurement results and the results obtained by laboratory gamma spectrometry measurements of the soil samples taken from the same locations

Comparative analyses of the measured radionuclides concentrations by the in-situ measurements and by sampling and laboratory measurements have been performed using the results obtained in the March 2007 campaign. These analyses showed an adequately agreement between the two methods for Cs-137 and radionuclides from natural series (e.g. Fig. 5 for Ac-228). However a systematic overestimation of the K-40 concentrations in soil samples was observed as compared with the in situ measurements (Fig. 6). This may be due to the presence of K-40 in building materials and in live organisms and this may bring to a variability of the background for this radionuclide.

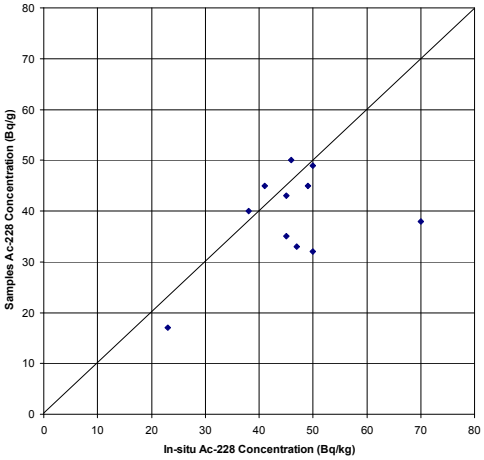

Fig. 5. Samples vs. in-situ Ac-228 Concentration

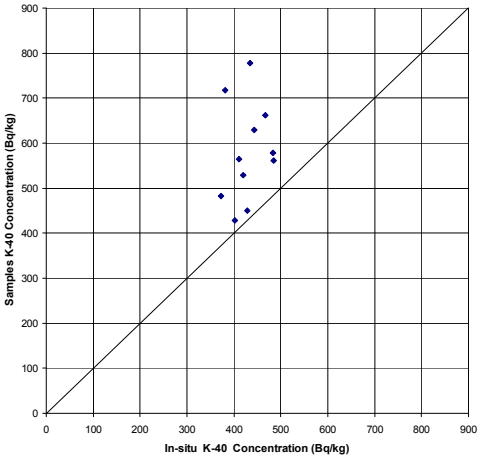

Fig. 6. Samples vs. in-situ K-40 Concentration

4. CONCLUSIONS

The dose rate measurement results are within the limits of the natural background. The average of the values recorded in monitoring locations from the vicinity of the Danube river bank and the average of the recorded values in locations situated on tributaries are close to each other. Variability of the results was limited enough as reported to the spatial distribution of the monitoring locations, showing that the gamma radiation background at the ground level for the entire monitored area was homogeneous.

The measurement results of the gamma emitting radionuclides concentrations in soil and sediment samples show a good equilibrium between the radionuclides from the natural series of Th-232 (Tl-208, Bi-212, Pb-212, Ac-228) and U-238 (Pb-214, Bi-214, Th-234). These concentrations are within the limits of the natural background (even in locations situated near Kozloduy NPP and Cernavoda NPP).

There is a good correlation between the gamma dose rate and its main natural radioactivity contributors (K-40 is the most important contributor – about 75% from global gamma emission).

The good agreement between measured dose rates and estimated dose rates based on the analysis of the soil and sediment samples may be interpreted as an argument for the representativeness of these samples, proving the uniformity of the gamma emitting natural radionuclides distribution in the vicinity of the sampling locations.

The comparative analyses of the results confirm the good quality of the in-situ measurements, underlining in the same time the importance of the correctness of the calibration hypothesis for the in-situ method and the sensibility of the results to deviation from these.

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