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ENVIRONMENTAL RADIOACTIVITY IN MANECIU RESERVOIR, PRAHOVA COUNTY, ROMANIA

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Abstract. The present work is related to the efforts of knowledge of the distribution of the different radioactive isotopes in the environment and their possible influences of the flora and fauna in different regions, as well as on the quality of the different aliments. For this study the rivers that assure the water for artificial reservoir created in village Maneciu, Prahova County. From the entrances in reservoir of the river Teleajen, have been collected separated probes, up to the depth of 2 m. The probes have been separated with a step of 5 cm and have been prepared using standard procedures. Each probe – in cylindrical plastic boxes with 100 g weight each – has been measured in the underground laboratory of NIPNE-HH from salt mine "Unirea" Slanic-Prahova. The preliminary experimental results indicate the absence of the signals related to the major nuclear accidents. An interesting result is related to the behavior of the radioactive isotope of 40K.

Key words: radioactivity background, rivers, reservoir, underground measurements, radioactive isotopes concentrations, nuclear accident memory.

1. INTRODUCTION

The Maneciu reservoir is located close to Maneciu town, Prahova County at the confluence of Teleajen and Telejenel rivers and has several functions such as: source of the drinking water for Ploiesti city and Valenii de Munte town, irrigation water, electric power plant and fishery. The reservoir was put into operation in 1984 and has a watershed of 247 km^2, the surface of the lake is 1.9 km^2.

The Teleajen river springs from Ciucas Massif (in the upper lane is called "Beer Creek"). It crosses 113 km until it flows in the Prahova river. In the Cheia depression receives as tributary the creek Tampa, and afterwards, at the exit of the mountains, an important tributary, the Telejenel river. Upper course of the

Telejenel river, upstream of the confluence with Castle Creek, is known as Stanei Creek.

The radioactive fallout which has occurred after the Chernobyl accident in April 26, 1986, was characterized by complex meteorological conditions. Consequently, the contamination of the soil surface was highly non-uniform. One cannot possibly assume that, even for small areas of a few square kilometers, the initial deposition was uniform and, subsequently, the variations observed in inventory were caused by migration with and/or within the soil. The soil erosion processes, associated with land degradation represents, for long term, a major problem for the environment.

Fig. 1 – Maneciu reservoir, image from Google Earth.

Besides nuclear accidents following which are released into the atmosphere large amounts of radioactive isotopes, to environmental radioactivity bring their contribution primordial radionuclides, and their descendants, whose half-life is comparable to the age of the Earth, of which the most known are 235U, 238U, 232 Th and 40 K. The contribution of these isotopes can vary depending on soil components, geographic location, their concentration in soil and geological and meteorological conditions [1]. Because the reservoir has other functions besides the flood protection such as drinking water sources for neighboring towns and villages, irrigation water sources for agricultural land, fishing, recreational areas, it is very important to determine the level of radioactive contamination, if it exists.

2. MATERIALS AND METHODS

Soil samples were collected on a vertical profile from Teleajen river. In the investigated area, a pit was dug from which the samples were taken from surface up to 200 cm deep with an increment of 5 cm.

Subsequently, the samples were dried, sieved and 100 g from each sample placed in cylindrical plastic boxes. The acquisition time varied between 73100 s and 344500 s.

The samples were measured in the Underground Laboratory of IFIN-HH from Slanic Prahova, located at a depth of 208 m (~ 610 meter water equivalent) in the Unirea salt mine.

Fig. 2 – The underground laboratory from Unirea salt mine, in Slanic-Prahova.

All samples were investigated by using a CANBERRA high resolution gamma ray system provided with an Extended Range Coaxial Ge Detectors (XtRa) (120% relative efficiency, energy resolution FWHM of 2.1 keV for the 1.33 MeV line of 60Co, HV 5000 V), U-type, housed in a radiation protection shield.

To achieve a significant background reduction, the shield, made of 5 cm of old Lead layer with < 25 Bq/kg of 210Pb and surrounded by 10 cm of recent Lead, was lined in its interior with 5 cm OFEC and OFHC Copper (99.99% pure Copper with 0.0005% oxygen content and 99.99% pure copper with no oxygen content respectively). In this way we have achieved a reduction of the natural background of about 3600 times with respect to unshielded ground gamma ray spectrometer over the energy range 40 keV–3000 keV [2, 3].

The gamma spectra were acquired by means of a Lynx Digital Signal Analyzer operated by Genie 2000 software (Canberra Genie 2000 software) while the efficiency calibration were performed by means of Canberra ISOCS-LABSOCS mathematical calibration software. The same software was used to determine the activity concentration of all radionuclides identified in the samples.

The most important radionuclides identified in samples taken from Teleajen River are 210Pb, 226Ra, 137Cs, 228Ac and 40K.

For each isotope identified in samples, their specific activity was calculated using the formula, [4]:

$$\Lambda = \frac{A_n}{m\,T_a\,\eta\,\varepsilon}, \tag{1}$$

where: Λ – specific activity [Bq/kg]; A_n – net area of analysed peak [counts]; m – mass [kg]; T_a – acquisition time [s]; η – decay probability and ε – efficiency.

The minimum detectable activity (MDA) was calculated using the formula [5]:

$$MDA = \frac{2.71+4.65\sqrt{A_t}}{m\,T_a\,\eta\,\varepsilon}, \tag{2}$$

where: MDA – minimum detectable activity [Bq/kg]; A_t – total area of the analyzed peak [counts]; m – mass [kg]; T_a – acquisition time [s]; η – decay probability and ε – efficiency.

In order to determine the correlation between the distributions of the radionuclides we calculated Pearson's correlation coefficients using [6] the following equation:

$$r_{xy} = \frac{\sum_{i=1}^{n}(x_i-\bar{X})(y_i-\bar{Y})}{\sqrt{\sum_{i=1}^{n}(x_i-\bar{X})^2\sum_{i=1}^{n}(y_i-\bar{Y})^2}}, \tag{3}$$

where: x_i, y_i – the two variables for which the correlation is calculated, $\bar{X},\bar{Y}$- average values of x_i and y_i.

In this sense we calculated correlation coefficients between 210Pb–226Ra, 40K–226Ra, 228Ac–226Ra and between the 210Pb$_{ex}$–137Cs. Lead excess (210Pb$_{ex}$) is derived by taking the difference between lead and radium activities in soil samples.

3. RESULTS AND DISCUSSIONS

The aquatic environment, one of the main components of the environment, has a special significance for human society. Water is of vital necessity for life on earth, for which the processes occurring in water are studied very closely in all their aspects. Aquatic pollution, classical or radioactive, influences virtually all living creatures which use this resource. The human society can be highly affected by water quality they use, especially if it is contaminated with radioactive products, [7, 8].

Erosion and sedimentation processes represent a major risk to terrestrial ecosystems by reducing agricultural production capacity and by reducing the surface of the reservoir, after which it can silt the reservoirs up to its disappearance. With the soil lost in the catchment area, significant amounts of plant nutrients and minerals needed are lost.

The investigated area received important amounts of 137Cs after Chernobyl accident from April 1986.

The erosion and sedimentation processes can be assessed using 137Cs and $210Pb_{ex}$ radionuclides as environmental tracers, [9, 10, 11, 12].

The samples were taken from the deltaic area of Teleajen river from Maneciu reservoir, from surface to 170 cm deep, in layers of 5 cm each. After conditioning, the samples were measured in the underground laboratory.

The data regarding Teleajen River are summarized in Table 1 and the correlation coefficients in Table 2. The graph representation of specific activities for 137Cs, 40K, 210Pb, 226Ra and 228Ac are shown in Figs. 3–7.

The depth distribution of 137Cs, 40K, 210Pb, 226Ra and 228Ac was determined experimentally in Maneciu reservoir.

In the investigated profile, the specific activity of 40K ranges from 478 Bq/kg to 664 Bq/kg, having an average activity of (478 ± 47) Bq/kg. For specific activity of 226Ra, the average activity is (31.3 ± 1.5) Bq/kg and individual values are from 28.2 Bq/kg to 34.0 Bq/kg. The specific activity of 228Ac are from 29.0 Bq/kg to 34.1 Bq/kg with an average value of (33.3 ± 1.1) Bq/kg. 137Cs has an average value of its specific activity of (7.7 ± 2.7) Bq/kg and individual values are from 2.7 Bq/kg to 13.6 Bq/kg. About 210Pb specific activity we observe that it has an average value of (33.0 ± 4.3) Bq/kg with individual values from 25.1 Bq/kg to 40.1 Bq/kg.

Following soil erosion and sedimentation processes that affects the soil, variations of the specific activity of 137Cs and $210Pb_{ex}$ in the soil occur. From Fig. 3 and Fig. 8 one can observe that the specific activities of 137Cs and $210Pb_{ex}$ have several maxima and minima situated practically at the same depth for both radionuclides. The correlation coefficient for 137Cs and $210Pb_{ex}$ is 0.90, which demonstrate that between these two variables are strongly correlated. The similar behavior of 137Cs and $210Pb_{ex}$ demonstrates that the sedimented soil in the investigated depth profile originates from direct erosion for low activities of both or from erosion of already sedimented soil for high activities. The erosion of sedimented soil could occur during heavy rains or fast snow melting.

Table 1

The depth distribution of radionuclides in Maneciu reservoir

No.	Depth (cm)	137Cs		210Pb		40K		226Ra		228Ac	
		Activity (Bq/kg)	Uncertainty (Bq/kg)	Activity (Bq/kg)	Uncertainty (Bq/kg)	Activity (Bq/kg)	Uncertainty (Bq/kg)	Activity (Bq/kg)	Uncertainty (Bq/kg)	Activity (Bq/kg)	Uncertainty (Bq/kg)
1	0–5	9.4	0.6	37.0	5.7	591	25	30.3	3,6	33.0	2,8
2	5–10	11.2	0.7	31.9	5.5	592	25	29.3	3,8	31.8	2,8
3	10–15	7.7	0.5	36.5	5.8	596	25	32.5	3,8	32.1	2,8
4	15–20	9.1	0.6	37.6	5.8	635	26	31.4	3,0	34.1	2,7
5	20–25	7.5	0.5	34.7	5.5	610	26	31.2	3,8	32.9	2,9
6	25–30	5.5	0.4	30.9	4.9	563	24	30.1	3,7	32.7	2,9
7	30–35	9.3	0.6	38.8	6.1	617	26	32.3	3,9	32.7	2,9
8	35–40	8.8	0.6	37.3	5.9	606	25	30.2	5,4	31.6	2,9
9	40–45	10.0	0.6	37.7	5.8	610	25	31.4	3,1	32.8	2,7
10	45–50	9.9	0.6	36.1	5.7	601	25	33.7	4,0	32.1	2,8
11	50–55	7.0	0.4	35.2	5.6	628	26	31.7	3,8	31.8	2,9
12	55–60	7.1	0.4	33.8	5.4	630	26	31.8	7,2	34.1	2,8
13	60–65	5.2	0.3	32.9	5.1	646	27	32.7	3,2	33.8	2,7
14	65–70	10.6	0.7	40.4	6.4	664	28	34.0	4,1	33.5	3,0
15	70–75	5.6	0.4	33.4	5.3	596	25	34.0	4,0	33.3	2,9
16	75–80	3.9	0.3	30.8	4.9	614	26	32.3	5,5	32.6	2,9
17	80–85	5.0	0.3	31.3	5.0	643	27	32.8	4,1	32.7	2,9
18	85–90	6.6	0.4	36.2	5.7	617	26	30.9	4,9	32.1	2,9
19	90–95	11.2	0.7	39.4	6.2	660	28	31.2	3,7	33.9	2,9
20	95–100	13.6	0.8	34.6	5.5	643	27	31.6	3,8	32.4	2,9
21	100–105	12.1	0.7	25.3	5.0	658	27	32.6	3,1	30.7	2,7
22	105–110	11.8	0.7	38.0	5.9	654	27	32.8	3,0	33.4	2,7
23	110–115	5.5	0.4	31.1	4.9	562	24	28.2	3,7	31.8	2,8
24	115–120	6.5	0.4	29.2	4.7	547	23	28.9	3,8	31.9	2,8
25	120–125	7.3	0.5	30.9	4.9	540	23	29.8	5,3	31.2	2,8
26	125–130	9.7	0.6	33.0	5.2	560	24	31.3	6,5	31.2	2,8
27	130–135	8.5	0.5	32.2	5.0	553	23	29.9	3,0	32.0	2,6
28	135–140	7.3	0.5	31.8	5.0	558	23	32.8	4,0	32.6	2,8

No.	Depth (cm)	137Cs		210Pb		40K		226Ra		228Ac	
		Activity (Bq/kg)	Uncertainty (Bq/kg)	Activity (Bq/kg)	Uncertainty (Bq/kg)	Activity (Bq/kg)	Uncertainty (Bq/kg)	Activity (Bq/kg)	Uncertainty (Bq/kg)	Activity (Bq/kg)	Uncertainty (Bq/kg)
29	140–145	5.6	0.4	29.4	4.7	509	21	30.4	4,5	30.6	2,8
30	145–150	3.5	0.2	25.1	4.0	478	20	28.5	3,4	29.0	2,8
31	150–155	4.1	0.3	27.1	4.4	552	23	30.9	3,7	31.2	2,8
32	155–160	4.4	0.3	27.5	4.4	562	24	30.2	3,7	32.5	2,9
33	160–165	4.4	0.3	26.8	4.3	541	23	31.1	3,9	31.9	2,8
34	165–170	5.4	0.3	27.1	4.4	524	22	30.5	3,8	31.2	2,8

Table 2

Correlation coefficients

Correlation between	Correlation coefficient
210Pb–226Ra	0.50
40K–226Ra	0.63
228Ac–226Ra	0.50
210Pb$_{ex}$–137Cs	0.78

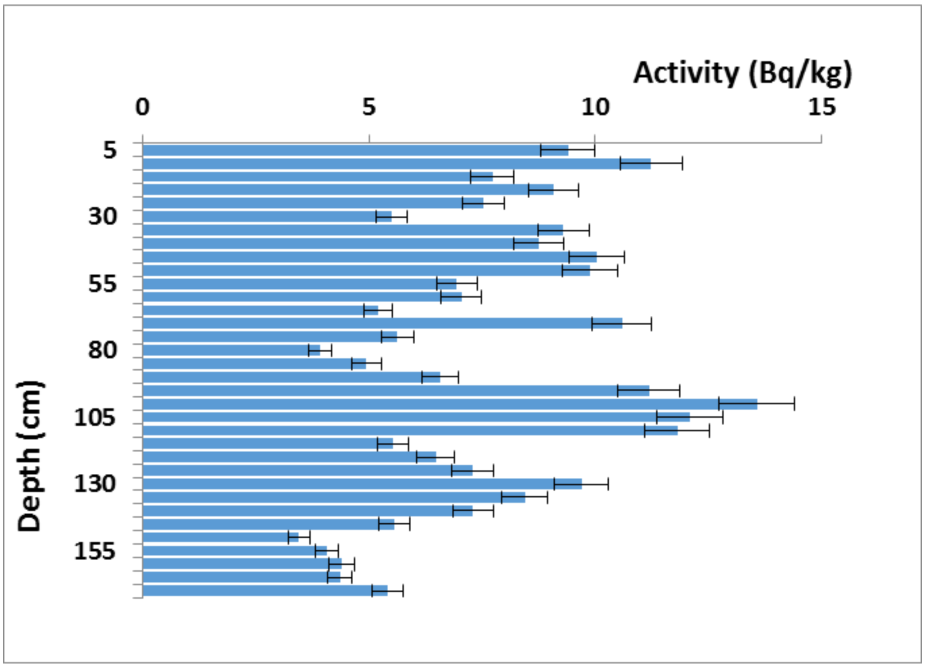

Fig. 3 – Depth profile of 137Cs in the Teleajen river.

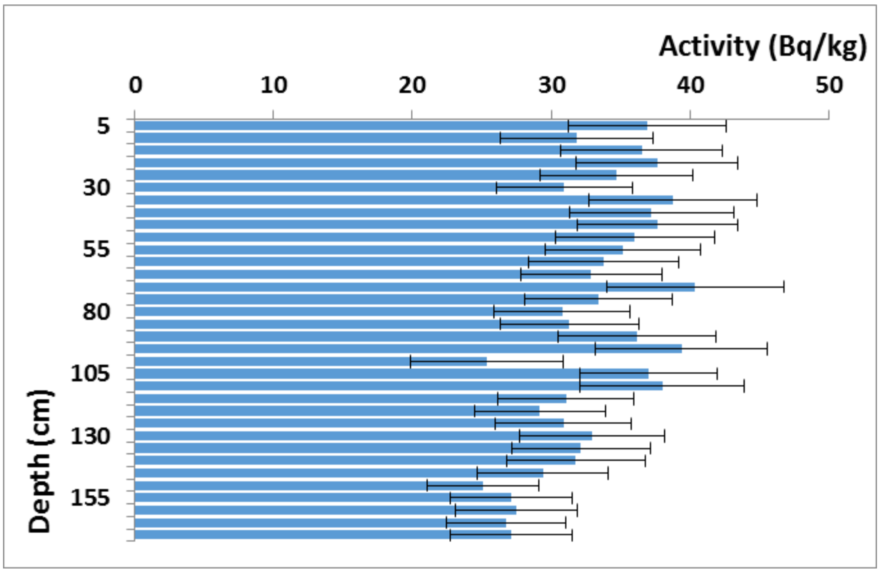

Fig. 4 – Depth profile of 210Pb in the Teleajen river.

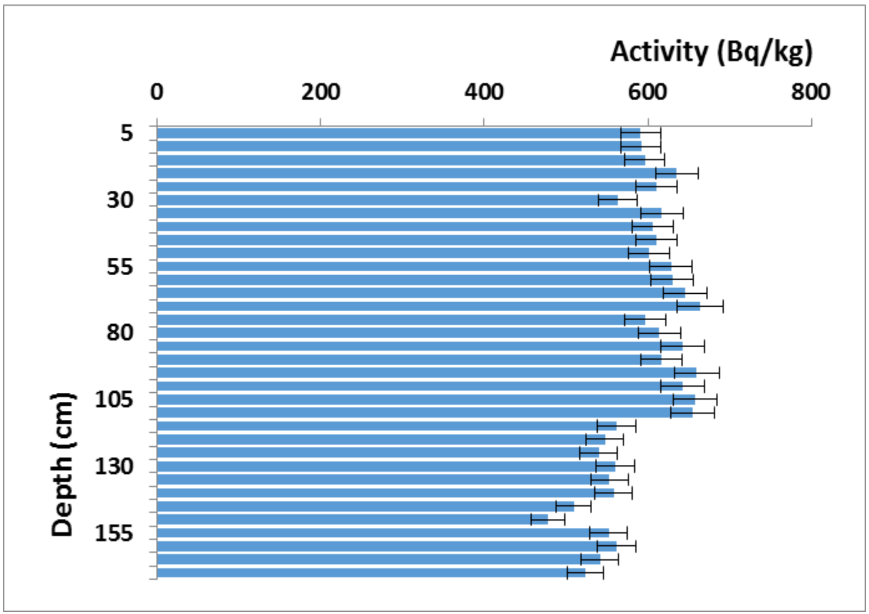

Fig. 5 – Depth profile of 40K in the Teleajen river.

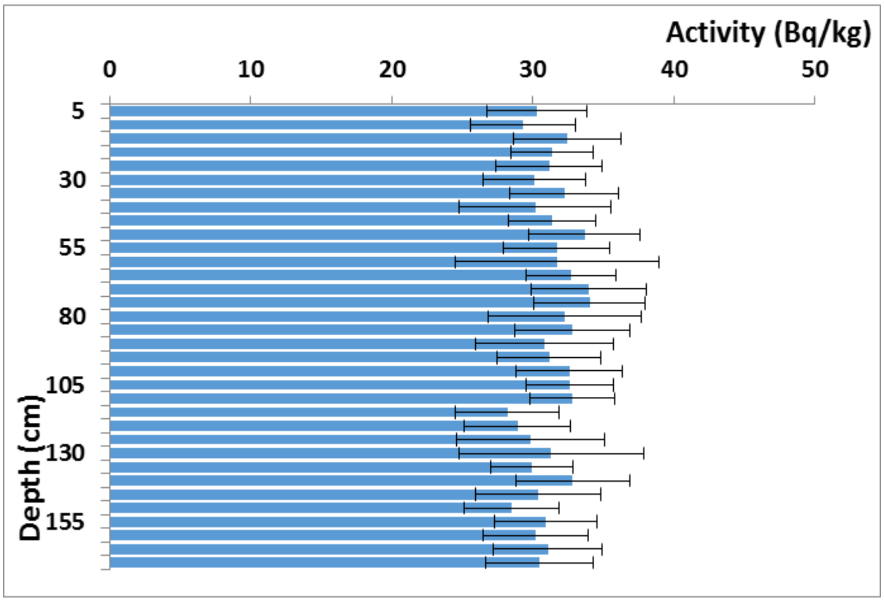

Fig. 6 – Depth profile of 226Ra in the Teleajen river.

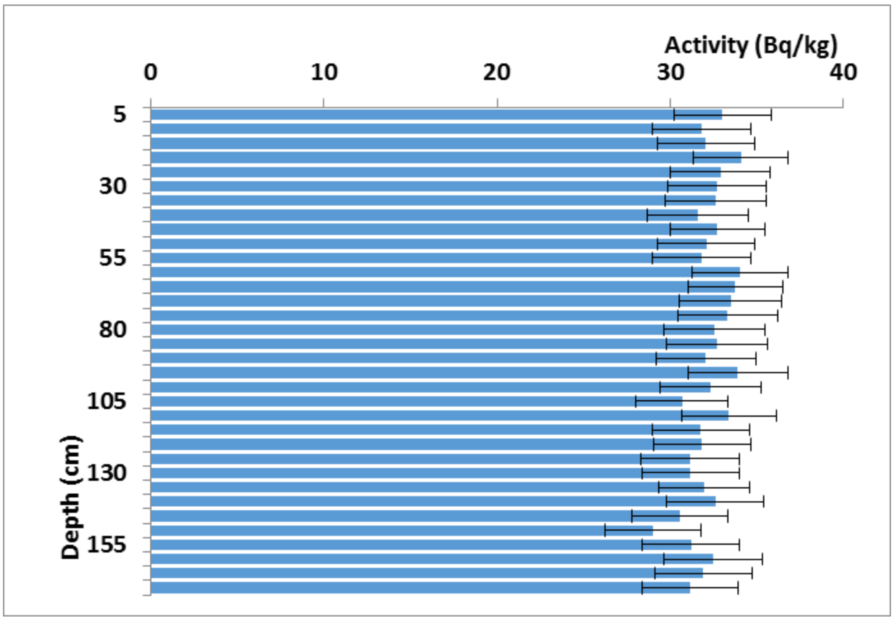

Fig. 7 – Depth profile of 228Ac in the Teleajen river.

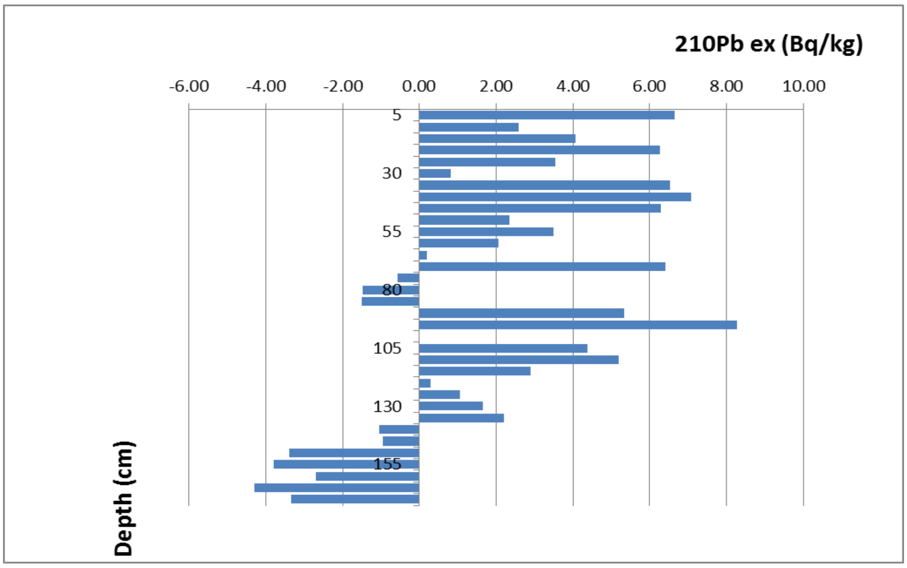

Fig. 8 – Profile of $^{210}Pb_{ex}$ with depth in Teleajen river.

In calculating the correlation coefficients 210Pb–226Ra and $210Pb_{ex}$–137Cs, we removed the corresponding sample values of 100–105 cm as 210Pb activity is unexpectedly low. In these circumstances, it was considered outlier value.

From Table 2 it can be seen that there is an acceptable correlation between 210Pb–226Ra, 40K–226Ra and, respectively, 228Ac–226Ra. Correlation coefficient of 0.78 between $210Pb_{ex}$ and 137Cs show that these two isotopes are well correlated.

In those circumstances, taking into account that the depth profile of 137Cs has several maxima and minima, this shows that the soil originating from the catchment has suffered a succession of erosion and sedimentation processes before being sedimented in the deltaic area of Teleajen river from Maneciu reservoir.

From experimental data, it can be seen that the activity of 137Cs are specific to the post Chernobyl deposition, up to a depth of 170 cm. In these conditions, we can appreciate that in Maneciu reservoir the sedimentation rate could be higher than 6 cm/year.

4. CONCLUSIONS

The investigated area is subject to surface erosion and subsequently sedimentation processes on the river bad. The investigated area received important amounts of Cs137 after Chernobyl accident from April 1986. The inventory of Cs137 in the sedimented eroded soil on the river bed varies in different places, even for the same reservoir.

The specific activity of 40K, 226Ra and 228Ac are specific for Romanian territory.

In the investigated profile, there is an acceptable correlation between 210Pb–226Ra, 40K–226Ra and, respectively, 228Ac–226Ra.

In the same profile, $210Pb_{ex}$ and 137Cs are well correlated, demonstrating that the sedimented soil originated from the watershed suffered a succession of erosion and sedimentation processes before its arrival in the deltaic area of Teleajen river.

It is rather difficult to evaluate the sedimentation rate and a supplementary investigation is necessary.

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REFERENCES

1. Vasilie N. S. (Florea), *Studiul Radioizotopilor Ambientali* (in Romanian), PhD Thesis, Faculty of Physics, University of Bucharest, 2012.
2. Romul Margineanu, Bogdan Mitrica, Ana Apostu and Claudia Gomoiu, Environ. Res. Lett. **6**, 034011 (2011).
3. R.M. Margineanu, Ana-Maria Blebea Apostu, Claudia-Maria Gomoiu, C. Costea, Delia Dumitras, Adriana Ion, O.G. Duliu, Journal of Environmental Radioactivity **138**, 72–79 (2014).
4. Knoll G. F., *Radiation Detection and Measurement*, 3rd Edition, John Wiley&Sons, 1999.
5. Gilmore G., *Practical Gamma Ray Spectrometry*, 2nd Edition, John Wiley&Sons, Ltd. 2008.
6. Viorica Postelnicu, Silvia Coatu, *Mica enciclopedie matematică* (translation in Romanian from “Kleine Enzyklopädie der Mathematik”), Editura Tehnică, București, 1980.
7. I. Ionita, R. Margineanu, Acta Geologica Hispanica **35**, *3–4*, 311–320 (2000a).
8. I. Ionita, R. Margineanu, C. Hurjui, Acta Geologica Hispanica **35**, *3–4*, 357–367 (2000b).
9. Rodway-Dyer S.J, Walling D.E, Journal of Environmental Management **91**, 1952–1962 (2010).
10. Deu. P., Walling D.E., Applied Radiation and Isotopes **69**, 717–726 (2011).
11. Paolo Porto, Desmond E. Walling, Antonina Capra, Soil & Tillage Research **135**, 18–27 (2014).
12. Blebea-Apostu A.M., Radulescu I., Margineanu R., Ionita I., Popescu I.V., Romanian Reports in Physics **64**, *1*, 211–220 (2012).