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Cs137 AND K40 IN SOIL AND WATER ALONG THE ROMANIAN SECTOR OF DANUBE RIVER

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ABSTRACT
The most important sources of artificial radionuclides from the Danube River in the last 70 years are the nuclear weapon tests in atmosphere and the Chernobyl accident. Cs-137 is one of the radiological most important long-lived radionuclides of anthropogenic origin, which has accumulated in the environment. This aspect was considered to be the most important for long-term radiological impact due to higher emissions, relatively long half-life and relatively high bioavailability. In 2015 we conducted campaigns of sampling soil and water from the Danube River between Bazias and Murighiol. Measurements of radionuclides in the samples were performed in the underground laboratory of IFIN-HH from Slanic-Prahova. The high resolution gamma-ray spectrometry laboratory is equipped with two HPGe detectors, produced by CANBERRA. The first detector has a relative efficiency of 22%, and the second detector has a 120% relative efficiency. The specific activity of 137 Cs in water samples from the Danube was generally below the limit of detection. For samples measured for very long time, more than 200000 s, the Cs137 activity in water was measurable, with values between 0.011 and 0.020 Bq/dm^3. Regarding the specific activity of K40, it was revealed in only two samples with values of 0.325 Bq/dm^3, respectively 0.699 Bq/dm^3 in all the other, it was below the detection limit. In soil, along the Danube River floodplain there are places with high values, ex. Hinova and adjacent areas, with values up to 140 Bq/kg, respectively Isaccea with values around 75 Bq/kg. In some points, Cs137 had values below 2 Bq/kg, otherwise they stood in the range 4-32 Bq/kg. The specific activity of K40 in the soil was in the range of 400 Bq/kg to 700 Bq/kg, with t.wo exceptions value of less than 400 Bq/kg and two higher than 700 Bq/kg.
Keywords: Cs137, K40, underground laboratory, soil, water, Danube River

1 INTRODUCTION

The Danube River is alongside the Carpathian Mountains and the Black Sea, one of the major components of the landscape, to which we define placing Romania on the continent. Grigore Antipa said about the Danube River that it is "the greatest treasure which nature has endowed our country".

The Danube River is the second longest rivers of Europe (after the Volga), the only European river that flows from west to east. It springs in the Black Forest, Germany below the Kandel peak, at an altitude of about 1240 m far lower than its tributaries from the Alps that spring from 2800-3000 m. The flow of Danube River at its spring varies between 4 and 10 m^3/s with losses in limestone mountains which it traverses. Its length, (Marinescu 1985; Gastescu and Stiuca 2008; Popp 1985), from its spring to the Black Sea is about 2860 km, and a river basin of 817 000 km^2, fig. 1, representing 8% of Europe.

The Danube is an important international river road, flowing through ten countries: Germany, Austria, Slovakia, Hungary, Croatia, Serbia, Romania, Bulgaria, Moldova and Ukraine and has tributaries in seven other countries. All ten countries through which the river flows are Member States of the IAEA (International Atomic Energy Agency) and passes through four state capitals: Vienna, Bratislava, Budapest and Belgrade.

In Romania the Danube is crossed by five bridges (Iron Gates dam 1 Giurgiu-Ruse, Fetesti-Cernavoda, Giurgeni - Vadu Oii and Calafat-Vidin) and three railroads (Giurgiu-Ruse, Fetesti-Cernavoda and Calafat-Vidin). Along the river, in Romania, there are port towns in Orsova, Drobeta Turnu Severin, Calafat, Corabia, Turnu Magurele, Giurgiu, Oltenita, Calarasi, Fetesti, Cernavoda and Harsova, Macin, Braila, Galati, Isaccea, Tulcea and Sulina, (Marinescu 1985).

Near Danube operates several nuclear facilities, among which the most important are: Packs NPP (Hungary), Kosloduy NPP (Bulgaria) and Cernavoda NPP (Romania), (Maringer 2000). These NPPs, and not only, can be potential sources of radioactive contamination of the environment.

We live with radiation. All environmental factors contain smaller or higher amounts of various radioactive isotopes, (Ion-Mihai 1998). Soil and water are two environmental factors that naturally contain radioactive isotopes.

Knowledge of environmental radioactivity level is required both during normal operation of nuclear facilities and in undesirable case of nuclear accident. Because nuclear power plants are considered as potential sources of radioactive pollution of the environment, it is imperative to execute a permanent control on radioactive contamination of the environment.

Figure 1. Danube River basin,
http://ec.europa.eu/regional_policy/archive/cooperation/danube/images/danube_topog.png (April 2016)

About a radionuclide, we say that it is natural when its existence is the result of natural processes without human intervention. In contrast a radionuclide is called artificial, when its existence is the result of human activities. Radioactive substances in the environment can be divided into two large groups after their origin. Depending on the origin, we are talking about natural or artificial radioactivity. Since its formation, the Earth has an inventory of radioactive isotopes. Part of that initial inventory not yet disintegrated is what exists now. As a result, natural radionuclides of this kind are those having a long half-life, comparable to the age of the Earth ($4.6 * 10^9$ years): U238 $T_{1/2} = 4.468 * 10^9$ years, Th232 $T_{1/2} = 1.4 * 10^{10}$ years, U235 $T_{1/2} = 7.9 * 10^8$ years and K40 $T_{1/2} = 1.28 * 10^9$ years. These natural radionuclides are considered of earthy origin.

By artificial radioactivity of the environment, we understand the presence in the environment of radionuclides which owe their existence to human activity. Artificial sources of radioactivity are: nuclear tests in the atmosphere, normal and abnormal functioning (accidental) of a nuclear facility, (nuclear reactor, nuclear fuel processing plant, radioactive waste storage). It should be emphasized that normally, the impact of all these activities is small compared to the natural radioactivity, however, in cases of accident or nuclear tests, the effects can be dramatic, see:
(https://en.wikipedia.org/wiki/Lists_of_nuclear_disasters_and_radioactive_incidents).

The release of radioactive substances into the environment, characteristic of nuclear explosion or a nuclear accident contains fission products and activation. Among the fission products, Cs-137 radionuclide is one of the relatively long half-life, 30.14 years, (Browne and Firestone, 1986). In case of nuclear accident or nuclear explosions in the atmosphere, significant amounts of Cs-137 are released, which can affect the environment in the long term, up to 300 years. Chernobyl nuclear power plant, located in Ukraine in 1986 had four nuclear reactors in operation. Following a test on April 26 of that year, was triggered the largest nuclear accident known until then, (OECD 1995). Comparable to the Chernobyl accident is the nuclear accident which occurred on 11 March 2011 in Fukushima, Japan, (Povinec 2013, Busby 2011).

Following the Chernobyl accident, huge amounts of radionuclides have been released into the atmospheres, which have affected the whole of Europe, Romania inclusively, (Paunescu et all, 1992, Margineanu at al. 1998, Mihai et al, 1998, Mihai et al, 1999). Since the beginning of the period of testing nuclear weapons in the atmosphere in years, 50's of last century, the radioactive contamination of the environment has been monitored and evaluated by national health authorities in terms of minimizing the health risks of the population, (Maringer 2000). Danube has also become an important objective for radio-monitoring and evaluation of artificial radioactivity.

EXPERIMENTAL METHODS

Gamma-ray spectrometry is used to study gamma radiation fields (γ) emitted by different radionuclides, for their identification. This method allows identification of natural and artificial radionuclides present in the analyzed material. Gamma-ray spectrometry is based on the fact that gamma radiations are emitted with discrete energy, well-defined, specific to emanating radionuclides.
A block diagram of a spectrometric system is shown in fig. 1

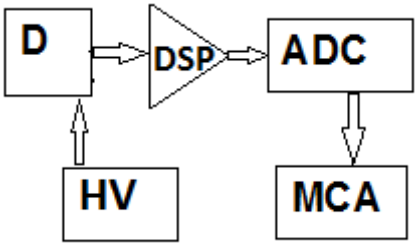

Figure 1. D – detector; DSP – Digital Signal Processor; HV – High Voltage; ADC – Analog to Digital Convertor; MCA – Multi Chanel Analyzer

The high resolution spectrometric systems used to measure the soil and water samples are equipped with HPGe detectors. In 2015 we carried out three campaigns for soil and water sampling from the Danube between Bazias and Murighiol to find out the current status of radioactivity of the Danube riparian area.

The sampling points are shown in Fig. 2 for area between Bazias and Zimnicea and in the Fig. 3 for the area between Giurgiu and Murighiol.

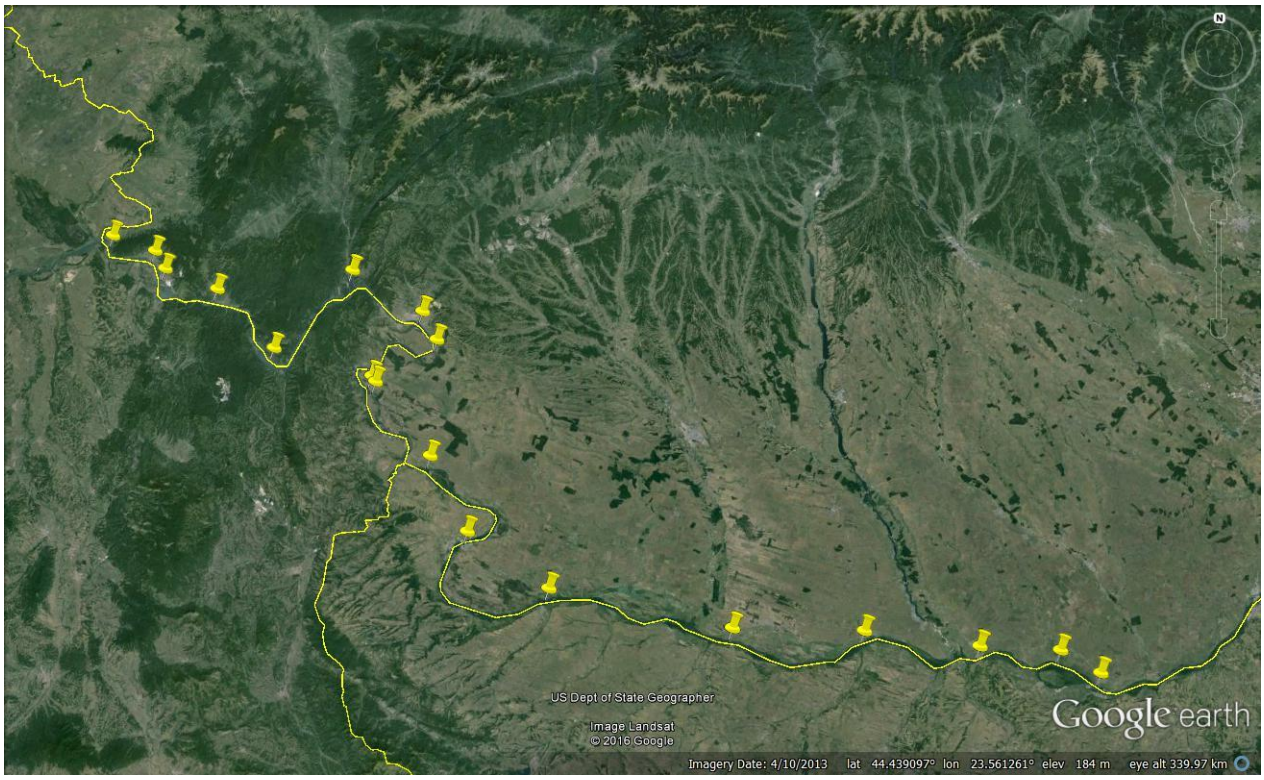

Figure 2. Sampling points from Bazias to Zimnicea

Figure 3. Sampling points between Giurgiu and Murighiol.

After conditioning, the samples were measured by high resolution gamma spectrometry in the IFIN-HH underground laboratory located in Unirea salt mine from Slanic-Prahova.

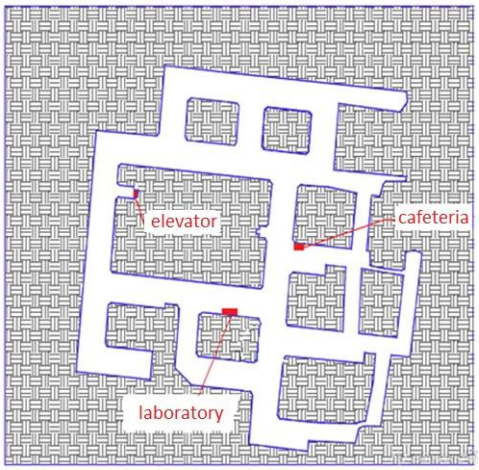

Figure 4. Unirea salt mine galleries plan and laboratory position

The laboratory is located underground at a physical depth of 208 m, its position as is shown in Fig. 4, is on one the main galleries on the wall opposite the main pillar. In the laboratory of high resolution gamma spectrometry are two HPGe detectors, produced by CANBERRA. The first detector has a relative efficiency of 22% and the second detector has a 120% relative efficiency.

For the first detector, radiation protection shield is made of three layers, the inner 2cm copper OFHC, then 5 cm of old lead-containing Pb210 below 25 Bq/kg and at exterior 10 cm recently lead. For the second detector, the shield is made of 3 layers all, but on the inside are 5 cm copper OFHC, then 5 cm of old lead-containing Pb210 below 25 Bq/kg and at exterior 10 cm recently lead. In these conditions, the radiation background measured inside the shield is 3600-4000 times smaller than the surface, the detection limit being reduced accordingly.

RESULTS AND DISCUSSION

The environmental radioactivity in Cernavoda NPP area was measured during a 10 years period, beginning with 1984, (Margineanu et all, 1988). In Cernavoda NPP influence area, Cs137 and K40 were measured in soil samples taken from semi-natural field. In 1984 and 1985, the Cs137 activity in soil has had values ranging from <0.6 Bq/kg to 13 Bq/kg. After Chernobyl accident, Cs137 in soil in this area shows

activities up to 89 Bq/kg, in Rasova sampling site in 1986. In the next years, with some fluctuations, the activity of Cs137 in soil decreased slowly. The activity of K40 in soil, in this period, was in the range 300-700 Bq/kg.

In another study regarding the environmental radioactivity, the Romanian sector of the Danube River for two years was monitored in 1989 and 1990, (Paunescu et all., 1992). Cs137 activity in the first 5cm of soil has had values up to 635 Bq/kg in Hinova and 376 Bq/kg in Tulcea. In the rest of investigated sites, the activity of Cs137 was less than 90 Bq/kg.

Cs137 in soil, in 2015, along the Danube River has areas with high values, ex. Hinova and adjacent areas, with values up to 140 Bq/kg and, respectively Isaccea, close to Tulcea with values around 75 Bq/kg, fig. 5, fig. 6 and table 1. In some places, Cs137 has values below 2 Bq/kg, otherwise there are in the range 4-32 Bq/kg.

In the present work, the highest activity of Cs137 in soil was found, practically in the same places, Hinova and Isaccea. It is important to mention that the activity of Cs137 measured in soil in 2015 in the above mentioned places, is less than the theoretical value given by the radioactive decay law.

The specific activity of K40 in the soil was in the range of 400 to 700 Bq/kg, two exceptions have values <400 Bq/kg and other two over > 700 Bq/kg. Similar values for K40 in soil were reported in 1998, (Margineanu et all. 1998).

The specific activity of Cs137 in water samples from the Danube River taken in the same places were soil was sampled was generally below the limit of detection. For samples measured on very long time >200,000 Cs137 activity in water was measured, with values between 0.011 and 0.020 Bq/dm^3 see table 2.

Regarding the specific activity of K40, it was revealed only two samples with values of 0.325 Bq/dm^3, respectively 0.699 Bq/dm^3 in all the other samples, it was below the detection limit, see table 2.

Place	**Cs137 Bq/kg**	**Uncertainty Bq/kg**	**K40 Bq/kg**	**Uncertainty Bq/kg**
Bazias	1.0	0.2	582	41
Pojejena	22.2	1.0	594	41
Moldova Veche	13.6	1.0	476	44
Liborajdea	11.1	1.0	621	52
Orsova	8.2	0.6	742	62
Simian	78.4	4.7	701	64
Hinova	139.4	11.3	503	44
Tiganasi	23.3	1.5	427	36
Iron Gates II	1.1	1.0	356	21
Garla Mare	31.9	2.3	540	49
Calafat	5.6	0.4	374	21
Rast	4.0	0.3	680	53
Bechet	9.1	0.7	617	58
Corabia	19.5	1.6	638	58
Turnu Magurele	9.4	0.8	815	67
Suhaia	24.9	2.1	608	51
Zimnicea	13.0	1.0	525	45
Giurgiu	4.7	0.4	469	34
Greaca	6.5	0.5	700	65
Oltenita	0.6	0.1	501	45
Bogata	9.4	0.6	666	23
Calarasi	1.4	0.1	543	16
Roseti-Coslogeni	9.0	0.5	605	21
Galdau	6.9	0.3	525	15
Borcea	4.4	0.2	485	14
Fetesti	7.0	0.2	568	16

Place	Cs137 Bq/kg	Uncertainty Bq/kg	K40 Bq/kg	Uncertainty Bq/kg
Facaeni	13.2	0.7	493	18
Giurgeni	18.4	0.6	503	15
Stanca Stancuta	14.6	0.7	654	22
Braila bac	9.1	0.6	543	19
Galati bac	11.2	0.4	579	17
Ostrov	6.6	0.3	623	18
Garliciu	8.2	0.5	553	20
Daeni	15.2	0.8	609	22
Peceneaga	7.6	0.3	524	15
Macin	4.9	0.4	531	20
Isaccea	74.6	2.4	478	14
Tulcea	18.2	0.8	489	17
Murighiol	3.0	0.2	423	13

Table 1. Activity of Cs137 and K40 in soil in Danube River Floodplain

Place	Cs137 Bq/dm^3	Uncertainty Bq/dm^3	K40 Bq/dm^3	Uncertainty Bq/dm^3
Moldova Noua	0.020	0.003	<0.39	
Iron Gates II	0.012	0.002	<0.19	
Giurgiu	0.012	0.003	0.33	0.08
Bogata	0.011	0.003	<0.25	
Isaccea	<0.02		0.70	0.07

Table 2. Activity of Cs137 and K40 water in several places in Danube

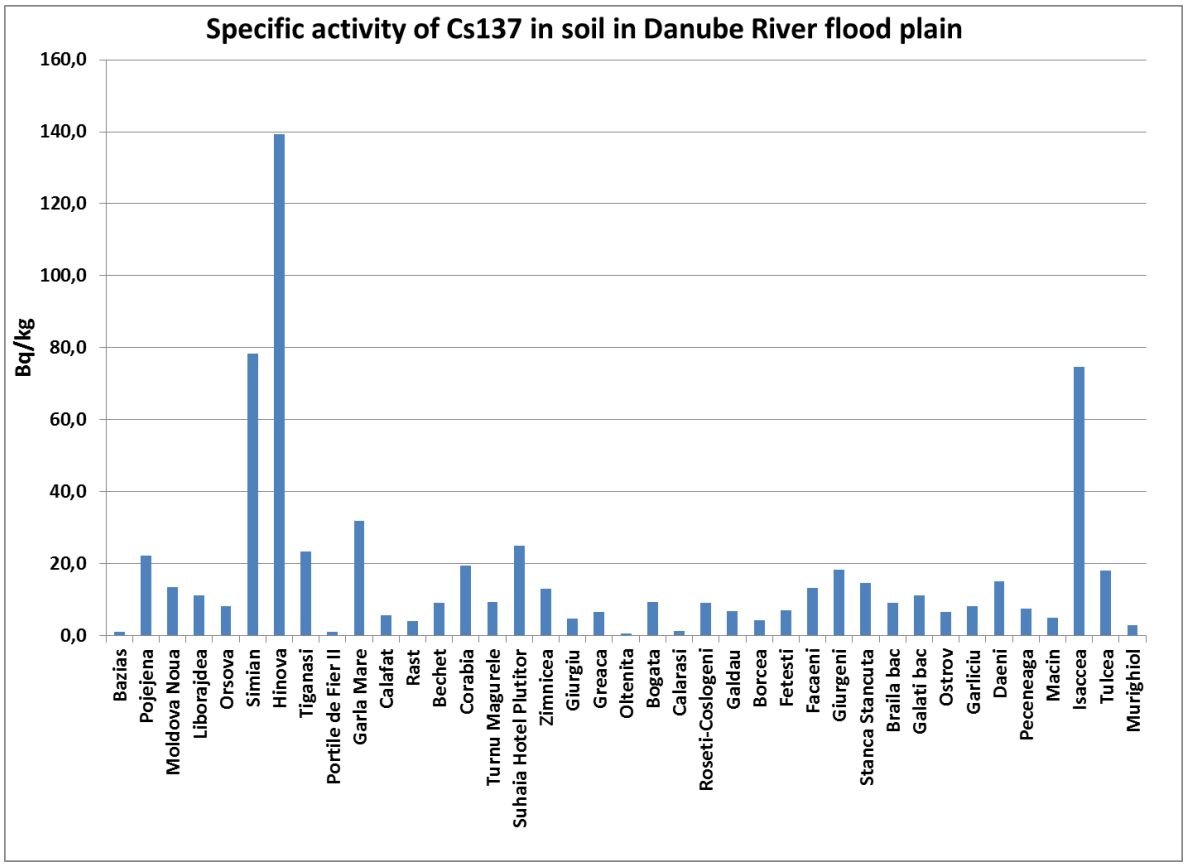

Figure 5. The specific activity of 137 Cs in the floodplain of the Danube River

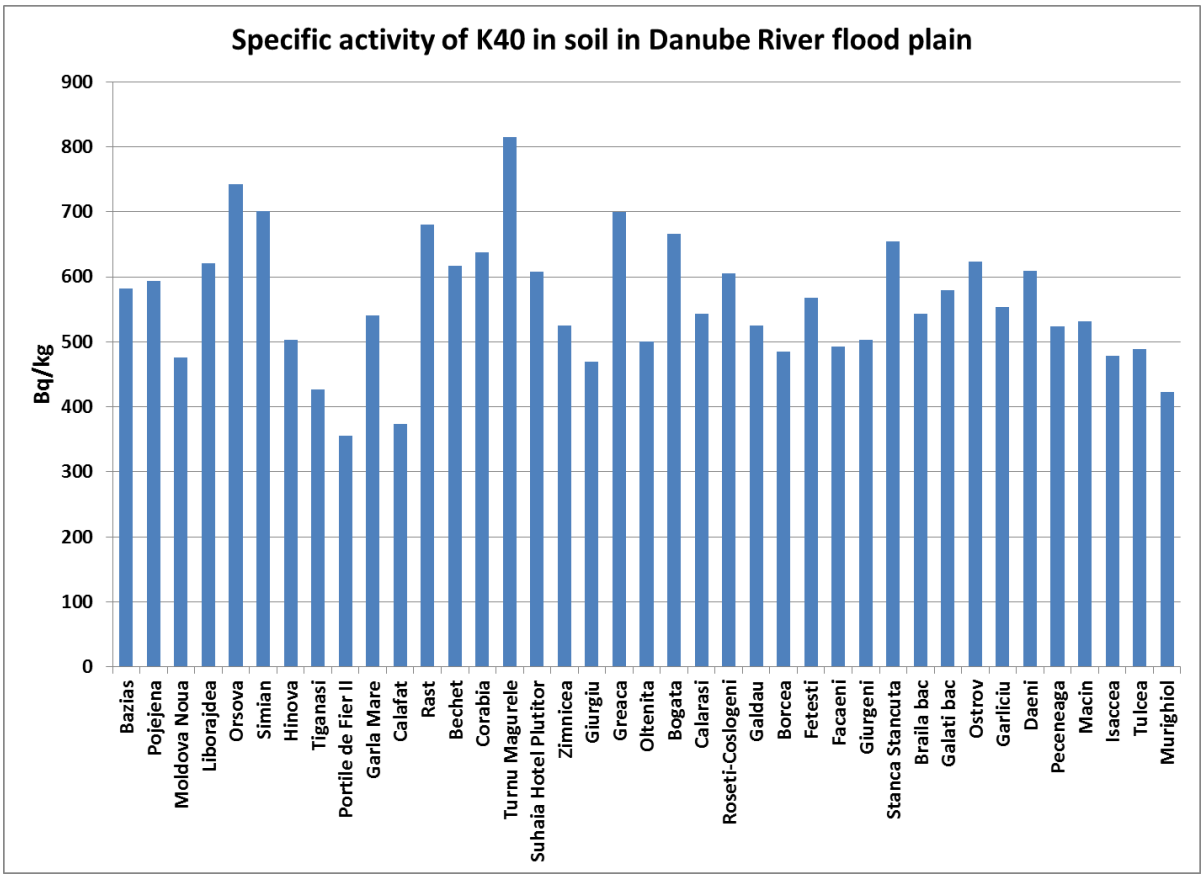

Figure 6. The specific activity of K40 in the floodplain of the Danube River

CONCLUSIONS

The specific activity of Cs137 in water samples from the Danube River was generally below the limit of detection. For few samples measured on very long time >200,000, Cs137 activity in water was measured, with values between 0.011 and 0.020 Bq/dm^3. Regarding the specific activity of K40, only two samples have values above minimum detectable activity, with values of 0.325 Bq/dm^3, respectively 0.699 Bq/dm^3 Cs137 in soil, along the Danube River has areas with high values, ex. Hinova and adjacent areas, with values up to 140 Bq/kg, respectively Isaccea with values around 75 Bq/kg. With few exceptions the rest of the values are in the range 4-32 Bq/kg.

The specific activity of K40 in the soil was in generally in the range of 400 Bq/kg to 700 Bq/kg with few exceptions.

The presence of Cs137 in Danube River environment demonstrates that the monitoring of radioactivity of this area is still a necessity.

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