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RECONSTRUCTION OF THE MONTENEGRO TERRITORY CONTAMINATION WITH $^{239+240}$PU ISOTOPES IN 1994

Abstract

On the basis of *in situ* gamma-spectrometric measurements of the ^{137}Cs activity concentrations in 1994, activity concentrations of $^{239+240}$Pu isotopes in soil on 47 locations in Montenegro have been estimated. A maximum measured ^{137}Cs activity in that time was 740 Bq/kg, and maximum estimated $^{239+240}$Pu activity – 14.8 Bq/kg. For the estimations, the activity ratio between these two isotopes ($^{239+240}$Pu/^{137}Cs) was used. This ratio has been determined recently, using alpha and gamma-spectrometric measurements (an average: 0.02, standard deviation: 0.007). The results showed that the sources of $^{239+240}$Pu and ^{137}Cs contamination in Montenegro are both the nuclear weapons testing and accident at Chernobyl.

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REKONSTRUKCIJA KONTAMINIRANOSTI TERITORIJE CRNE GORE IZOTOPIMA $^{239+240}$PU 1994. GODINE

Izvod

Na osnovu *in situ* gama-spektrometrijskih mjerenja koncentracija aktivnosti ^{137}Cs 1994. godine procijenjene su aktivnosti izotopa $^{239+240}$Pu u zemljištu na 47 lokacija u Crnoj Gori. Maksimalna izmjerena vrijednost aktivnosti ^{137}Cs tada je bila 740 Bq/kg, a maksimalna procijenjena aktivnost $^{239+240}$Pu – 14.8 Bq/kg. Za procjene je korišćen odnos aktivnosti ova dva izotopa ($^{239+240}$Pu/^{137}Cs), utvrđen nedavno alfa i gama-spektrometrijskim mjerenjima (srednja vrijednost: 0.02, standardna devijacija: 0.007). Rezultati su pokazali da su izvori $^{239+240}$Pu i ^{137}Cs u Crnoj Gori testiranja nuklearnog naoružanja i havarija u Černobilju.

INTRODUCTION

Man-made radiation sources ^{137}Cs and $^{239+240}$Pu have come in the environment of Montenegro as a result of the global fallout from nuclear weapons testing (from 1945 to 1980), and the Chernobyl reactor accident in 1986. The first systematic survey of ^{137}Cs activity in the soil of Montenegro was performed using *in situ* γ-spectrometry in 1994 (i.e., eight years after the Chernobyl accident) [1]. On the other hand, the first measurements of $^{239+240}$Pu activity in Montenegro soil have been performed recently (by alpha-spectrometry, in the Radiation and Nuclear Safety Authority STUK – Helsinki, Finland, under the contract between the STUK and Montenegrin Academy of Sciences and Arts; in the frame of the Academy's project *Reconstructing level of contamination of the Montenegrin territory by plutonium of Chernobyl origin*), and the $^{239+240}$Pu/^{137}Cs activity ratio has been determined to be 0.02, with a standard deviation of 0.007. It is important to point out that ^{239}Pu and ^{240}Pu are not usually distinguished in environmental measurements, and this mixture ($^{239+240}$Pu) contains about 60 % of ^{239}Pu in terms of activity.

As it is known, for estimating level of territory contamination with plutonium isotopes some correlation coefficients can be applied, such as correlations between activity of plutonium isotopes and activities of ^{144}Ce, ^{103,106}Ru, ^{137}Cs, etc. [2–6].

For applying this method, it is necessary to have results of gamma-spectrometry of soil samples (i.e., ^{137}Cs activity in a sample) as well as to know the $^{239+240}$Pu/^{137}Cs activity ratio. Then, it is possible to estimate $^{239+240}$Pu activity concentration in the same sample. By applying this ratio, it is even possible to reconstruct a contamination level at any moment in the past (but after the Chernobyl accident), and that is done in the present study. By knowing ^{137}Cs activity concentrations in surface soil in Montenegro in 1994, a level of soil contamination with $^{239+240}$Pu isotope in that period has been reconstructed, and the results are presented and discussed here.

This approach is based on an assumption that the $^{239+240}$Pu/^{137}Cs activity ratio was practically the same in 1994 as nowadays, which follows from some previous research showing that in the middle of the nineties these two chemically different elements (from the tests of nuclear weapons) were fixed together in the soils in the same ratio as it is analyzed over many years ago [7]. The same follows from our research of activity ratios for these two isotopes ($^{239+240}$Pu/^{137}Cs) in different soil layers (0–5, 5–10 and 10–15 cm), where practically the same correlation coefficients have been found.

EXPERIMENTAL

In situ gamma-spectrometry

A survey of ^{137}Cs concentration in the soil of Montenegro was performed eight years after the Chernobyl accident (in 1994) using the method of *in situ* γ-spectrometry, with portable γ-spectrometer, consisting of HPGe detector (n-type, beryllium window, 100 cm^3 active volume, 1.95 keV FWHM at 1332 keV), 4k-multichannel analyzer and lap-top computer. The measuring sites and the municipality centers in Montenegro are shown in Fig. 1.

The ^{137}Cs measuring locations in 1994 were chosen according to their geological, pedological and other characteristics. A regular 15 x 20 km sampling grid covered the whole territory of Montenegro, and one measuring site was selected in each of the grid rectangles. In general, each measured cesium activity was representative for a larger area.

Figure 1. Locations of measurements : ◊ measuring site, ○ municipality center

Results and discussion

The measurements performed in 1994 showed that the ^{137}Cs activities ranged from 3.3 to 74 kBq/m^2, with a mean of 16.05 kBq/m^2, and median of 13.7 kBq/m^2. The highest contamination of 74 kBq/m^2 was found at the site in the mounting Sinjajevina, at the altitude of 1702 m (marked with **A** in Fig. 1). The measurements revealed that the ^{137}Cs contamination, in the first approximation, followed topographical map of Montenegro, increasing with altitude of the region [1].

The ^{137}Cs activity concentrations on 47 locations measured by *in situ* gamma-spectrometry in 1994, and estimations of $^{239+240}$Pu activity concentration in that period for the same locations, are given in Table 1 and Figs. 2 and 3, respectively, while statistics of measurements and estimations – in Table 2.

Table 1. Results of 137Cs measurements and $^{239+240}$Pu estimations for soil in Montenegro in 1994.

	Location	Measured ^{137}Cs activity concentration, [Bq/kg]	Estimated $^{239+240}$Pu activity concentration, [Bq/kg]
1	Jakupov grob (Boljanići)	359±37	7.18±0.74
2	Gornja Rudnica (Pljevlja)	174±19	3.48±0.38
3	Trsa (Piva)	444±37	8.88±0.74
4	Potkokot (Bobovo)	629±74	12.58±1.48
5	Drljino brdo (Kosanica)	444±37	8.88±0.74
6	Kozička Rijeka	444±37	8.88±0.74
7	Seljani (Piva)	252±26	5.04±0.52
8	Pašina voda (Durmitor)	444±37	8.88±0.74
9	Donja Dobrilovina (Tara river)	222±22	4.44±0.44
10	Glibavac (Tomaševo)	326±33	6.52±0.66
11	Mokri lug (Bistrica)	285±30	5.70±0.60
12	Dubočke (Banjani)	115±11	2.30±0.22
13	Duga (Nikšić)	266±26	5.32±0.52
14	Kruševice (Šavnik)	292±22	5.84±0.44
15	Bunar Smrdan (Sinjajevina)	740±74	14.80±1.48
16	Marića Luka (Mojkovac)	263±26	5.26±0.52
17	Bioča (Berane)	45±4	0.90±0.08
18	Bašča (Rožaje)	326±33	6.52±0.66
19	Vilusi	289±22	5.78±0.44
20	Kuside (Nikšić)	314±30	6.28±0.60
21	Kuta (Nikšić)	252±26	5.04±0.52
22	Kodža (Međuriječje)	85±7	1.70±0.14
23	Mateševo (Bare Kraljske)	481±37	9.62±0.74
24	Šekular	159±15	3.18±0.30
25	Kozare (Rožaje)	363±33	7.26±0.66
26	Dragalj	270±26	5.40±0.52
27	Čevo	141±14	2.82±0.28
28	Ždrebaonik (Danilovgrad)	44±4	0.88±0.08
29	Piperska Rijeka (Radunovići)	81±7	1.62±0.14

	Location	Measured ^{137}Cs activity concentration, [Bq/kg]	Estimated $^{239+240}$Pu activity concentration, [Bq/kg]
30	Veruša	155±15	3.10±0.30
31	Martinovići (Gusinje)	252±26	5.04±0.52
32	Krašići (Tivat)	37±4	0.74±0.08
33	Bajice (Cetinje)	133±11	2.66±0.22
34	Draževina (Kruse)	56±4	1.12±0.08
35	Pikalja (Cijevna)	104±11	2.08±0.22
36	Bečići (Budva)	63±7	1.26±0.14
37	Boljevići (Crmnica)	48±4	0.96±0.08
38	Pothum (Tuzi)	59±4	1.18±0.08
39	Čeluga (Bar)	26±4	0.52±0.08
40	Curovići (Ostros)	37±4	0.74±0.08
41	Gornji Štoj (Ulcinj)	15±2	0.30±0.04
42	Ilino brdo (Pljevlja)	200±19	4.00±0.38
43	Budimlja (Berane)	100±11	2.00±0.22
44	Hotel „Durmitor” (Žabljak)	215±22	4.3±0.44
45	Motel „Lokve” (Berane)	30±4	0.6±0.08
46	Hotel „Bjelasica” (Kolašin)	107±11	2.14±0.22
47	Hotel „Sveti Stefan”	24±3	0.48±0.06

Table 2. Statistics of Cs measurements and Pu estimations.

	Minimum	Maximum	Mean	Standard deviation	Median
^{137}Cs [Bq/kg]	15	740	217	168	200
$^{239+240}$Pu [Bq/kg]	0.3	14.8	4.3	3.4	4

A cumulative ^{137}Cs and $^{239+240}$Pu activity concentration on each location is presented in Fig. 4.

As abovementioned, the $^{239+240}$Pu/^{137}Cs activity ratio for Montenegro soil is estimated to be 0.02 with standard deviation of 0.007, which is significantly lower than its mean value of 0.41 in Jordanian soil samples [8], slightly lower than 0.037±0.007 found in *Rokkasho*, Japan [9], but similar to some measurements in Europe shortly before the Chernobyl accident, where values around 0.018 have been found [10, 11]. On the other hand, it is higher than 4.8×10^{-3} in Northern Switzerland, where calculations included fractions of ^{137}Cs in soil samples which were contamina-

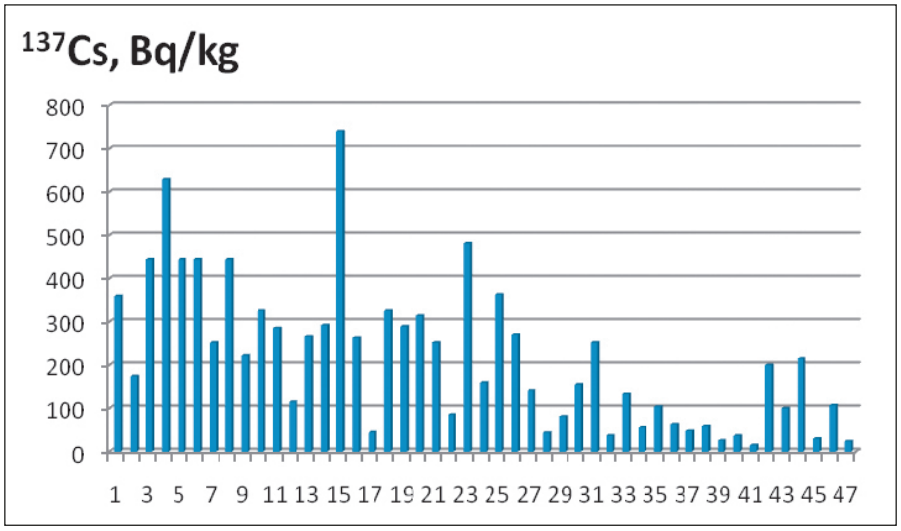

Figure 2. ^{137}Cs activity concentration in 1994 (47 locations in Montenegro)

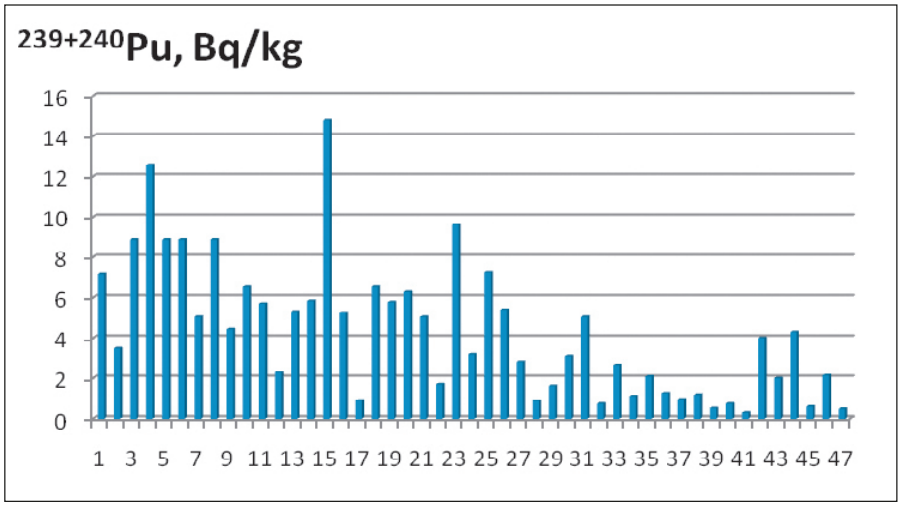

Figure 3. $^{239+240}$Pu activity concentration in 1994 estimated for 47 locations in Montenegro

ted by fallout products from previous (40-years-ago) atmospheric nuclear bomb testing and from the 1986 Chernobyl accident [12].

The average $^{239+240}$Pu/^{137}Cs activity ratio in Montenegro soil corresponds to values of the same ratio for some sediments (for example, 0.0155

to 0.0411, with a mean of 0.0215 – in sediment core of Lake Chenghai, SW China [13], 0.02–0.04 – in bottom sediments of southern Baltic Sea [14], etc.), as well as to the annual activity ratio obtained in analyzing delivery of transuranic elements by rain to the Mediterranean Sea (0.023 – in rain samples collected at Monaco in the end of the seventies [15]).

The measurements in 1994 showed that ^{137}Cs activity concentrations in the soil of Montenegro ranged from 15 (Gornji Štoj – Ulcinj) to 740 (Bunar Smrdan–Sinjajevina) Bq/kg, with a mean of 217 Bq/kg and standard deviation and median of 168 and 200 Bq/kg, respectively (Tables 1 and 2, Fig. 2). The $^{239+240}$Pu activity concentration has been estimated to range from 0.3 to 14.8 Bq/kg, with a mean, standard deviation and median of 4.3, 3.4 and 4 Bq/kg, respectively (Tables 1 and 3, Fig. 3). At the same time, for example, ^{137}Cs activity concentrations measured in Ukraine, in the two soil samples, were 960 and 1210 Bq/kg [16], and the activity ratios ^{238}Pu/$^{239+40}$Pu and $^{239+240}$Pu/^{137}Cs suggested that the proportion of cesium radioisotopes and ^{239,240}Pu in the soil attributable to the Chernobyl accident was approximately 100 % and 10–20%, respectively [16].

As mentioned previously, $^{239+240}$Pu and ^{137}Cs were deposited on the surface soil in Montenegro during two time periods (in the sixties – nuclear weapons testing, and in 1986). The corresponding fractions originating from the global fallout in the sixties and from the Chernobyl fallout in 1986, cannot be resolved in this case. That can be achieved by measuring the isotopic ratios ^{238}Pu/$^{239+240}$Pu and ^{134}Cs/^{137}Cs, because these ratios were significantly different in the fallout from these two sources. As in many European countries (where ^{137}Cs was deposited in considerable amounts by the Chernobyl fallout in 1986) the global fallout can be separated from Chernobyl fraction by means of the ^{134}Cs isotope. However, there are no data about ^{134}Cs in Montenegro soil, and this will no longer be possible to find out due to the ^{134}Cs short half-life. Previous research of ^{134}Cs/^{137}Cs ratio in some European regions (e.g. Central Europe [12]) indicated that more than 80 % of surface bound ^{137}Cs can be attributed to fallout of aerosols from the Chernobyl in 1986. It is not possible to determine precisely this percentage for Montenegro soil because there are no historical (pre-Chernobyl) data available. That is also reason why determined ^{137}Cs/$^{239+240}$Pu activity ratio cannot be used for developing an equation as a mean of resolving the plutonium which came from the Chernobyl accident from that which came from global fallout

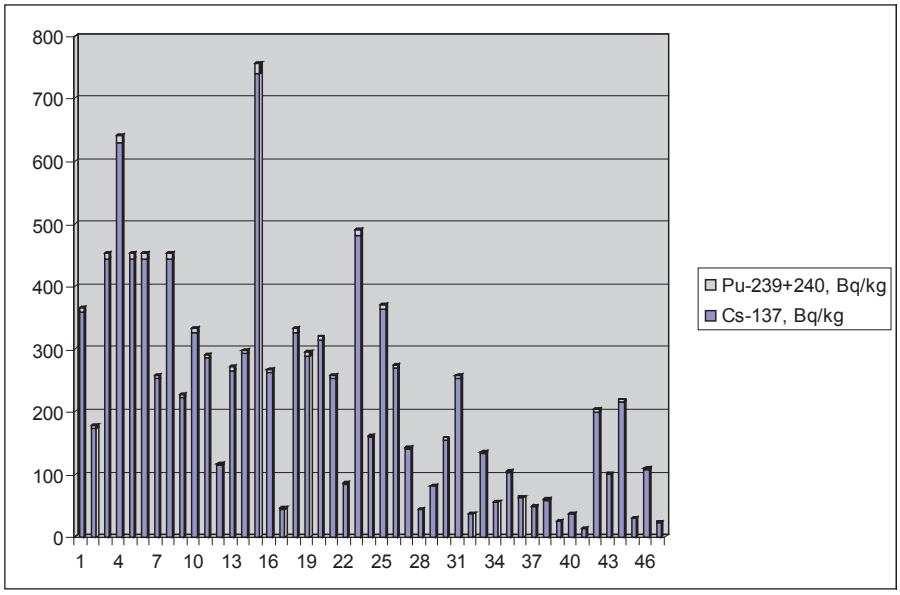

Figure 4. A cumulative (^{137}Cs and $^{239+240}$Pu) activity for all 47 locations

(as it is performed in the case of plutonium in attic dust and soil from Nevada and Utah – came from the Nevada Test Site fallout and came from global fallout [17]).

Finally, it is important to emphasize that, although it is usually assumed that $^{239+240}$Pu originates in most European countries essentially only from the global fallout [18], the determined $^{239+240}$Pu/^{137}Cs activity ratio for Montenegro soil differs from the global fallout average (related to atmospheric nuclear weapons testing), and indicates soil contamination from other sources, in this case from the Chernobyl accident. In a further research, the ^{238}Pu/^{137}Cs and ^{238}Pu/$^{239+240}$Pu activity ratio for Montenegro soil will be analyzed, and that could help in obtaining more information about the Chernobyl component in the total contamination level, not only in 1994, but also nowadays.

CONCLUSIONS

The present study determines contamination level of the Montenegro soil with ^{137}Cs and $^{239+240}$Pu in 1994, i.e., their activity concentrations on 47 locations. The ^{137}Cs activity ranged from 15 to 740 Bq/kg, whilst $^{239+240}$Pu activity – from 0.3 to 14.8 Bq/kg.

It follows from the results that the source of $^{239+240}$Pu and ^{137}Cs in Montenegro is deposition from both the atmospheric nuclear tests and Chernobyl accident. For obtaining precise information about the Chernobyl accident contribution to the Montenegrin territory contamination, the ^{238}Pu/^{137}Cs and ^{238}Pu/$^{239+240}$Pu activity ratio should be also analyzed.

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