Radioisotopes in ground soil *terra rossa* samples from the island of Mljet

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INTRODUCTION

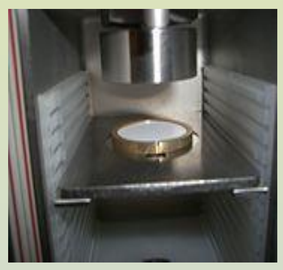
- The main goal was screening of the current state and determination of target radioisotopes activity ratio in *terra rossa* soil samples
- The target radioisotopes were ^{90}Sr, alpha emitters (^{241}Am, ^{239,240}Pu), as well as naturally occurring and anthropogenic gamma emitters (^{137}Cs)
- The radioisotopes distribution in surface soil layers is of the interest because of the possible uptake by plants
- *Terra rossa* is a typical soil developed at carbonate bedrock of the island
- Previous studies [1,2] of ^{137}Cs and naturally occurring gamma emitters activities in the *terra rossa* soil indicated both the nuclear weapon testing and Chernobyl contamination

MATERIALS AND METHODS

- The soil was sampled from the Mljet National Park (the north-west part of the island of Mljet, Croatia)

- The samples were prepared by oven-drying at 105 ^{o}C during 24 h and subsequently grinded
- Samples for gamma spectrometry determination were prepared in Marinelli beakers and analysed on a HPGe gamma detector (ORTEC)
- After gamma spectrometry measurement, every sample was kept in the burnout furnace at 600 °C for 24 h

- Alpha emitters were determinatinated from ashed and digested samples
- Concentrated nitric acid was used for digestion
- Am i Pu are mutually separated on column filled with Anion Exchange Resin (8 moldm^{-3} nitric acid)
- Am preconcentrated with CaC_2O_4 precipitation and isolated by an additional column filled with TRU resin (both resins Eichrom Technologies)
- Am and Pu were co-precipitated with NdF_3, filtered through a 0.2 μm (25 mm) pore size polysulfone membrane filter (Tuffryn HT-200, Pall Corporation) and determined by the ORTEC alpha spectrometer 576A equipped with ULTRATM ion implanted silicon detectors (active area 600 mm^2).



- ^{90}Sr was determined from ashed and digested samples
- 8 moldm^{-3} nitric acid
- Separation on column filled with AnaLig Sr-01 resin (IBC Advanced Technologies) [4]
- Cherenkov counting on TRI CARB 3100 TR (PerkinElmer)

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PRELIMINARY RESULTS

Table 1. Massic activities (Bq kg^{-1}) of anthropogenic radioisotopes in the soil samples

Soil	A (^{137}Cs) Bq kg^{-1}	A (^{90}Sr) Bq kg^{-1}	A (^{239,240}Pu) Bq kg^{-1}	A (^{241}Am) Bq kg^{-1}
1	46.3 ± 3.7	66.0 ± 5.9	0.382 ± 0.033	0.004 ± 0.001
2	18.9 ± 1.5	92.8 ± 8.3	0.505 ± 0.040	0.529 ± 0.051
3	24.7 ± 2.0	133.6 ± 11.9	0.598 ± 0.051	0.061 ± 0.007
3A	123 ± 9.8	209.9 ± 18.7	1.545 ± 0.121	0.376 ± 0.041
4	185 ± 15	330.1 ± 29.4	2.876 ± 0.268	0.286 ± 0.025
4A	140 ± 11	174.6 ± 15.5	3.912 ± 0.32+	0.429 ± 0.069
5	25.5 ± 2.0	198.7 ± 17.7	0.200 ± 0.014	0.957 ± 0.091
6	118 ± 9	204.0 ± 18.2	2.946 ± 0.279	0.243 ± 0.022
7	27.1 ±2.2	125.5 ± 11.2	0.602 ± 0.005	0.208 ± 0.019
8	80.2 ± 6.4	186.2 ± 16.6	2.129 ± 0.190	0.566 ± 0.06
9	82.6 ± 6.6	136.6 ± 12.2	1.750 ± 0.181	0.005 ± 0.001
10	0.5 ± 0.1	131.3 ± 11.7	0.107 ± 0.008	0.007 ± 0.001
11	70.1 ± 5.6	166.4 ± 14.8	1.066 ± 0.102	0.128 ± 0.011

- Activities of ^{90}Sr were higher than activities in soil samples collected from the exclusion zone (< 30 km) of the Fukushima Daiichi NPP presented in work of Sahoo and col. who obtained ^{90}Sr activity in range from (3.0 ± 0.3 to 23.3 ± 1.5) Bq kg^{-1} [5]

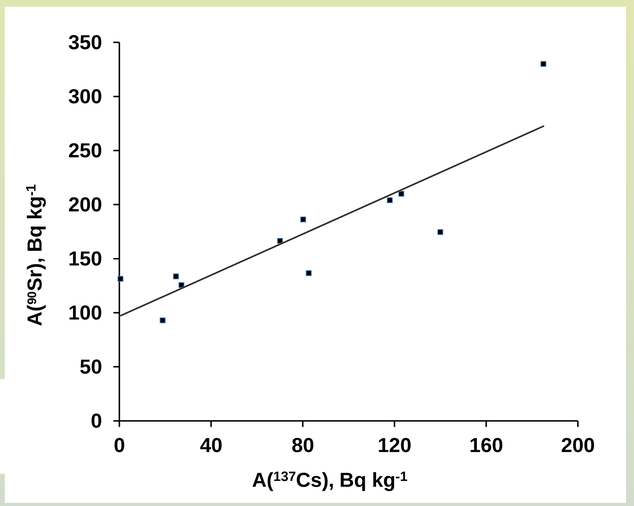
Table 2. Massic activities (Bq kg^{-1}) of naturally occurring radioisotopes in the soil samples

Soil	A (^{40}K), Bq kg^{-1}	A (^{226}Ra), Bq kg^{-1}	A (^{228}Ac),Bq kg^{-1}
1	4.79E+02 ± 1.55E+01	2.54E+02 ± 1.28E+01	7.73E+01 ± 3.45E+00
2	3.82E+02 ± 1.21E+01	1.87E+02 ± 9.37E+00	5.09E+01 ± 2.92E+00
3	5.65E+02 ± 1.61E+01	3.32E+02 ± 1.34E+01	5.77E+01 ± 3.69E+00
3A	5.95E+02 ± 1.42E+01	3.30E+02 ± 1.31E+01	5.14E+01 ± 3.09E+00
4	4.89E+02 ± 1.75E+01	4.32E+02 ± 1.40E+01	6.18E+01 ± 3.46E+00
4A	4.99E+02 ± 1.81E+01	4.87E+02 ± 1.50E+01	6.87E+01 ± 3.77E+00
5	5.03E+02 ± 1.47E+01	1.75E+02 ± 9.24E+00	6.32E+01 ± 2.98E+00
6	3.81E+02 ± 1.36E+01	1.16E+02 ± 1.03E+01	5.65E+01 ± 3.23E+00
7	5.38E+02 ± 1.67E+01	3.45E+02 ± 1.23E+01	6.83E+01 ± 2.65E+00
8	5.30E+02 ± 1.56E+01	4.22E+02 ± 1.32E+01	6.24E+01 ± 3.02E+00
9	3.93E+02 ± 1.32E+01	2.47E+02 ± 1.07E+01	6.29E+01 ± 3.39E+00
10	6.01E+02 ± 1.49E+01	2.03E+02 ± 9.69E+00	8.16E+01 ± 2.40E+00
11	4.49E+02 ± 1.68E+01	2.21E+02 ± 1.29E+01	8.11E+01 ± 2.99E+01

- Measured activities of ^{40}K were found within the previously reported ranges in *terra rossa* soils in Croatia and Slovenia, while ^{137}Cs activities were found higher [1,2]. The activity ratio between ^{90}Sr, ^{239,240}Pu and ^{137}Cs can be used to predict activities of these hardly detectable radioisotopes.

Table 3. Activity ratio of studied radioisotopes

^{137}Cs /^{90}Sr	**0.417 ± 0.080**
^{239,240}Pu/^{137} Cs	0.019 ± 0.006
^{239,240}Pu/^{90}Sr	0.008 ±0.002



- Figure 1. The correlation between the ^{90}Sr and ^{137}Cs massic activities in the soil samples is moderate

- After Chernobyl accident ^{137}Cs/^{90}Sr ratio was 8.5 in the fallout [6]. During IAEA mission in 1999 on testing site in Algeria ^{137}Cs/^{90}Sr activity ratio of 3.2 was reported for collected samples [7] The preliminary ^{137}Cs/^{90}Sr ratio obtained in this research, suggests conclusion that determined radioisotopes mainly originated from weapon testing deposition.

Conclusion

- **The presence of the anthropogenic radioisotopes in the collected samples**
- **Significantly lower ^{137}Cs/^{90}Sr activity ratio in *terra rossa*, in comparison with majority of previously reported for soils, was found**
- **Further research, especially ^{137}Cs / ^{90}Sr activity ratio is required for comprehensive conclusions**
- **Collected data with the interpretation of obtained results should be a baseline for the estimation of the impact for the possible future radioactive contamination on this protected area**

Acknowledgment

This study was supported by the KEGA 002UK-4/2016. Authors wish to thank the Ministry of Science and Education, the Republic of Croatia for M. Nodilo's salary during scientific stay at Comenius University and Prof. RNDr. P. Rajec for help with analysis of gamma spectra.