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Radiological impact of an active quarry in the Papuk Nature Park, Croatia

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Papuk Nature Park, unlike most similar parks and preserves in the world, contains active quarries. Quarries dig stone from the ground, creating dust and exposing deeper, potentially more radioactive layers. Since the forest trails in the Park lead right up to the quarries, we believed it was important to determine the radiological impact of the quarries on the Park environment. We measured ambient dose rate equivalent $H^*(10)$ and sampled moss at 26 Park locations along two of four quarries, along the road between them, and near Lake Orahovac, a very popular tourist destination close to the quarries. Moss is a standard bioindicator of exposure to heavy metals, including radionuclides. Using-gamma ray spectrometry we determined the activity concentration of ^{137}Cs and of representative naturally occurring radionuclides – ^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th, and ^{40}K – in sampled moss. $H^*(10)$ at selected locations was similar to the background $H^*(10)$ measured continuously all over Croatia. The ranges of measured activity concentrations of ^{137}Cs and naturally occurring radionuclides in moss did not differ significantly from other parts of Croatia and nearby countries.

KEY WORDS: ^{137}Cs; ambient dose rate equivalent $H^*(10)$; gamma ray spectrometry; moss; naturally occurring radionuclides

Papuk is the highest mountain of eastern Croatia. In 1999, an area of Papuk was designated a nature park (1), and in 2007, Papuk Nature Park (PNP) was designated a Global Geopark by the United Nations Educational, Scientific and Cultural Organization (UNESCO) (2). In the eastern part of the PNP, there is the Radlovac Quarry complex consisting of four quarries close together: Hercegovac, Oršulica, Brenzberg-Točak, and Žervanjska. Hercegovac and Oršulica are sources of dolomite (sediment rock), while Brenzberg-Točak and Žervanjska provide diabase (volcanic rock) (Figure 1). While the quarries are not attractive to PNP visitors, they are very close to Lake Orahovac, a very popular tourist destination on the edge of the PNP. In addition, popular hiking trails lead through the forest right up to and around the quarries.

Quarries are considered a potential source of increased radioactivity, as stone digging can bring material containing increased radioactive content to the surface (3, 4). Research into potentially negative radiological impact of quarries has been focused on the exposure of quarry workers to ionising radiation (5–7) and on radioactive content analysis of quarry products (8–24). However, research of radiological effects of a quarry on its surroundings is not common. Quarries are often situated in solitary areas, even though some become tourist destinations and nature preserves as part of the remediation process, such as the Quarry Nature Park in Canada (25), Park Hall Country Park and Hume Quarry (26), Helsby Quarry Woodland Park (27), Quarry Park & Nature Preserve Stearns County, Minnesota (28), and Portland Quarries Nature Park (29, 30).

Considering that the Radlovac Quarry is part of the PNP and very close to major tourist and hiking routes, we wanted to ascertain that there was no adverse radiological effect on the close environment. To do that, we measured ambient dose rate equivalent $H^*(10)$ at 24 locations around and between two quarries and at two locations near Lake Orahovac. We also measured radionuclide content in moss collected from these locations using gamma ray spectrometry.

MATERIALS AND METHODS

We selected 24 locations along the Brenzberg-Točak and Žervanjska quarries and the road between them and two additional locations by Lake Orahovac, a couple of kilometres away (Figure 2). The locations were selected based on the availability of moss. All the locations except the two by Lake Orahovac were on forest trails.

Sampling and $H^*(10)$ measurements took place in April and May of 2018. At each location we registered the coordinates, measured ambient dose rate equivalent $H^*(10)$ and sampled moss in order to determine radionuclide content using gamma ray spectrometry. We did not consider investigating the closest quarry to Lake Orahovac, Hercegovac, as it was in the process of

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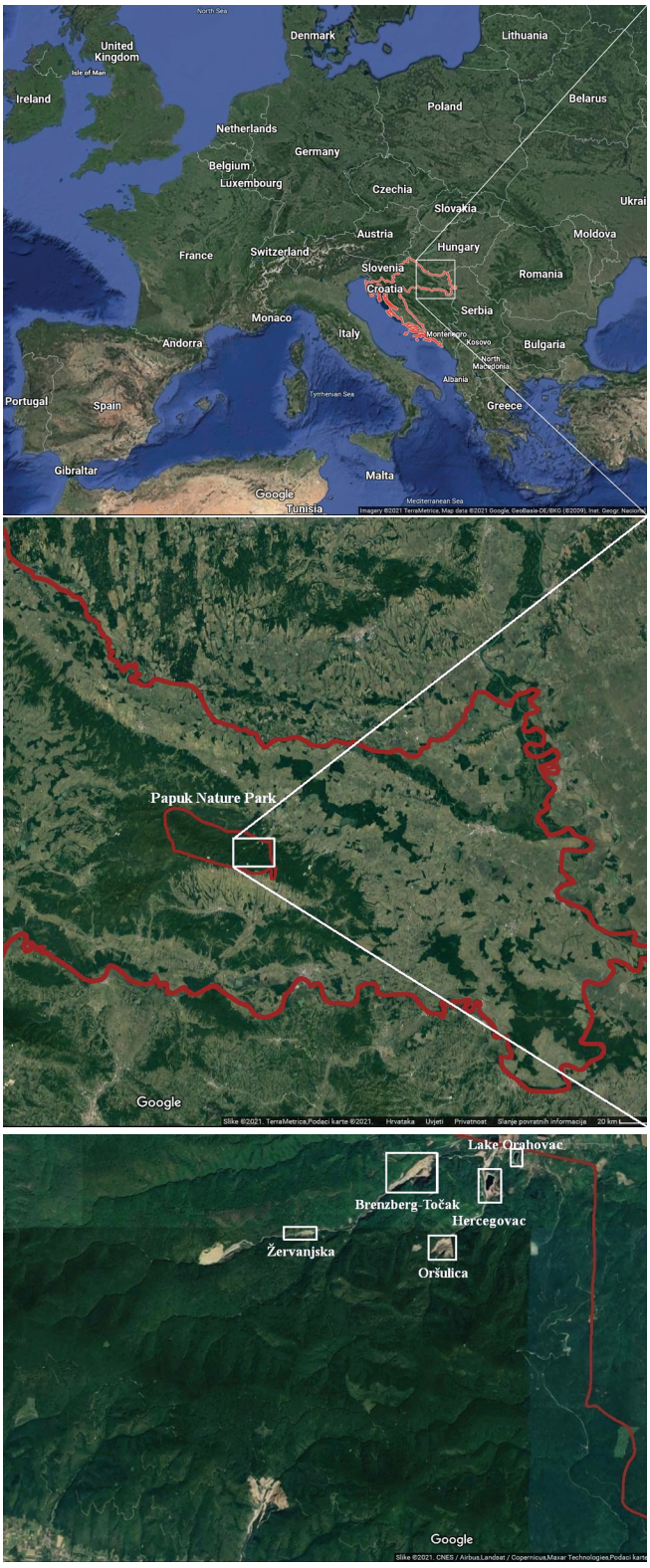

Figure 1 Location of the PNP within Croatia and locations of quarries Hercegovac, Oršulica, Brenzberg-Točak, and Žervanjska, as well as Lake Orahovac in the PNP (Source: Google Maps)

remediation. Instead, we selected the next two closest ones connected by a road.

*H**(10) measurements were taken with the RDS-31 S/R Multi-purpose Survey Meter (San Ramon, CA, USA) (31) at each location at the height of 1 m above the ground in no less than five three-minute gamma dose rate measurements within a radius of five meters from the central location to cover the whole area where moss was sampled. The survey meter was calibrated by the manufacturer ahead of measurements using the reference ^{137}Cs source and reference dose rate of 3 mSv/h.

While *H**(10) values are a solid indicator of radiological impact on the environment and people, true radiological exposure can only be assessed with a full characterisation of gamma-emitting radionuclides in the area.

Mosses are organisms that efficiently accumulate different elements from the environment and are often used as bioindicators of pollution (or its absence) with metals (32–36), including radioactive uranium, thorium, and caesium (37–40) or with potassium (37). Without a developed root system to absorb nutrients form the ground, mosses predominantly absorb nutrients and pollutants from the air (40–42). A recent research (42) suggests that under certain conditions, which include high availability of dust in the area and relatively low precipitation, the main source of nutrients and pollutants is not deposition from the atmosphere, but deposition of local dust.

Moss was collected, cleaned from dead leaves, soil, and other detritus, and packed into 5 L plastic bags for transport. Depending on the quantity available at each location, moss was collected within the radius of up to 5 m from the registered coordinates to ensure at least 1 L of moss for each sample after drying. Moss was dried to a constant mass in the laboratory and then packed and sealed in standard 1 L Marinelli beakers and left undisturbed for 30 days to establish secular equilibrium in the ^{238}U decay chain. We measured activity concentrations of selected radionuclides (naturally occurring radionuclides and ^{137}Cs) using the ORTEC gamma-ray spectrometry system (Advanced Measurement Technology, Inc., Oak Ridge, TN, USA), which uses a high purity Ge (HPGe) coaxial GMX-type detector (relative efficiency of 74.2 % and peak full width at half maximum of 2.24 keV, all at 1.33 MeV ^{60}Co). Energy and efficiency were calibrated using certified calibration sources manufactured by the Czech Metrology Institute (Jihlava, Czech Republic), and corrections for true coincidence made using the EFFTRAN program (43). Calibration source was a known mixture of radionuclides dissolved in H_2O packed into the same geometry (Marinelli beaker) that was used for sample measurements.

Each sample was measured for 80,000 seconds. The following peaks were analysed: 46.5 keV (^{210}Pb), 63.3 keV and the double peak at 92.4 and 92.8 keV (^{238}U), 351 keV and 609 keV (^{226}Ra calculated as ^{214}Pb and ^{214}Bi average), 662 keV (^{137}Cs), 911 keV (^{232}Th), and 1461 keV (^{40}K).

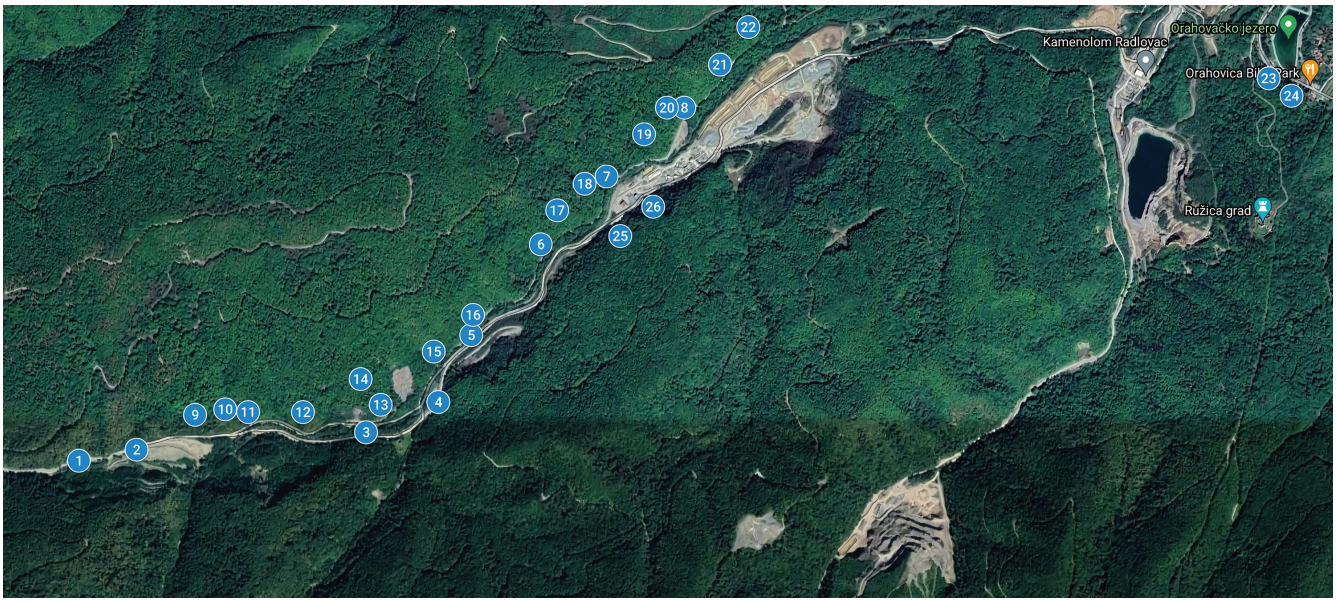

Figure 2 Locations of measurements near Žervanjska quarry (1, 2, 9), near Brenzberg-Točak quarry (7, 8, 19–22, 25–26), near the road between those quarries (3–6, 10–18) and near Lake Orahovac (23–24) (Source: Google Maps)

Table 1 *H**(10) measurements at 26 PNP locations around the Radlovac Quarry

Location	**1**	**2**	**3**	**4**	**5**	**6**	**7**
Latitude	45°30'12"	45°30'13"	45°30'15"	45°30'19"	45°30'27"	45°30'37"	45°30'45"
Longitude	17°48'09"	17°48'19"	17°48'58"	17°49'10"	17°49'16"	17°49'27"	17°49'38"
*H**(10) (μSv/h)	0.10±0.03	0.12±0.05	0.10±0.03	0.10±0.05	0.10±0.04	0.05±0.02	0.10±0.04
Location	**8**	**9**	**10**	**11**	**12**	**13**	**14**
Latitude	45°30'53"	45°30'17"	45°30'18"	45°30'17"	45°30'17"	45°30'18"	45°30'21"
Longitude	17°49'52"	17°48'29"	17°48'34"	17°48'38"	17°48'47"	17°49'00"	17°48'57"
*H**(10) (μSv/h)	0.14±0.05	0.11±0.03	0.11±0.03	0.12±0.03	0.10±0.03	0.09±0.03	0.11±0.03
Location	**15**	**16**	**17**	**18**	**19**	**20**	**21**
Latitude	45°30'25"	45°30'29"	45°30'41"	45°30'44"	45°30'50"	45°30'53"	45°30'58"
Longitude	17°49'09"	17°49'16"	17°49'30"	17°49'35"	17°49'45"	17°49'49"	17°49'58"
*H**(10) (μSv/h)	0.10±0.02	0.09±0.03	0.10±0.03	0.08±0.03	0.10±0.03	0.12±0.03	0.10±0.03
Location	**22**	**23**	**24**	**25**	**26**		
Latitude	45°31'03"	45°30'57"	45°30'55"	45°30'38"	45°30'42"		
Longitude	17°50'02"	17°51'30"	17°51'34"	17°49'41"	17°49'46"		
*H**(10) (μSv/h)	0.10±0.02	0.10±0.03	0.09±0.03	0.12±0.03	0.08±0.03		

RESULTS AND DISCUSSION

Ambient dose rate equivalents *H**(10) ranged between 0.05 μSv/h and 0.14 μSv/h, which is within the background range measured in Croatia (44, 45).

Moss radionuclide measurements are presented in Table 2. Table 3 shows the averages for the locations grouped as follows: along the Žervanjska quarry (locations 1, 2, 9; group 1), along the Brenzberg-Točak quarry (locations 7, 8, 19–22, 25–26; group 2), along the road between these two quarries (3–6, 10–18; group 3), and by Lake Orahovac (23, 24; group 4) (Figure 2). Moss samples collected by Lake Orahovac showed, on average, lower activity concentrations of naturally occurring radionuclides than samples collected along and between the quarries, save for ^{137}Cs. Samples collected along the road showed higher ^{137}Cs and lower ^{40}K activity concentrations than samples collected along the quarries. Samples collected along the Žervanjska quarry had higher ^{40}K, ^{232}Th, and ^{238}U activity concentrations than samples collected at other locations.

Table 2 Activity concentration measurements in moss collected at 26 PNP locations around the Radlovac Quarry

Location	Activity concentration (Bq/kg)					
	^{40}K	^{137}Cs	^{232}Th	^{238}U	^{226}Ra	^{210}Pb
1	409±20	2.50±1.5	29.2±5	35.8±10	15.5±1	241±35
2	222±10	12.1±0.9	13.8±2	24.8±6	20.4±1	303±20
3	357±20	35.0±2	12.9±3	18.5±8	18.5±1	607±50
4	379±10	2.47±0.7	24.6±3	39.1±10	37.3±2	233±30
5	178±10	25.9±1	12.5±2	17.8±6	19.2±2	778±40
6	170±10	8.17±1	9.99±3	17.0±13	5.54±0.4	363±30
7	150±9	24.1±1	9.57±2	12.6±5	11.6 ±0.9	654±30
8	300±10	5.56±0.6	5.92±1	8.50±4	6.18±0.7	672±30
9	203±10	25.4±2	15.1±3	21.6±11	4.72±0.6	903±60
10	247±20	11.8±1	15.9±3.5	37.8±10	18.2±2	996±80
11	167±9	7.11±1	8.98±2	12.0±5	13.2 ±0.9	800±30
12	173±10	11.2±1	10.7±2	11.0±6	8.04±0.6	897±40
13	213±10	20.9±2	13.5±3	14.2±7	30.0±2	289±45
14	144±10	22.4±2	10.2±3	<20	3±0.3	603±70
15	153±10	13.8±1	11.0±2	<5	4±0.5	779±70
16	178±10	16.1±2	10.7±3	11.5±8	27.8±3	498±50
17	222±20	24.6 ±2	14.3±4	17.4±6	21.8±2	405±60
18	199±10	25.8±2	13.3±3	17.6±10	11.5±1.5	620±40
19	189±10	24.5±2	8.71±3	24.0±20	12.2±1	625±60
20	184±10	12.6±1	10.1±2	18.0±7	19.2±2	754±40
21	213±10	15.3±1	9.07±3	12.6±6	4.15±0.4	564±40
22	332±10	7.65±1	11.5±2	16.3±12	29.9±2	418±60
23	232±10	25.7±2	6.49±3	<10	2±0.2	303±50
24	198±10	35.0±2	7.84±3	<10	<2	571±50
25	437±20	4.40±1	22.1±5	26.2±10	6.37±0.5	536±70
26	301±10	0.53±0.5	17.59±3	24.6±7	26.5±1.5	310±30
Range	**150–437**	**0.53–35.0**	**5.92–29.2**	**<DL–39.1**	**<DL–37.3**	**233–996**
Average	**236**	**16.2**	**12.9**	**19.9**	**15.1**	**566**

DL – detection limit, the lowest quantifiable value

To put these numbers in a context, Table 4 shows a comparison with findings from other studies in the same geographic area where possible.

Activity concentrations of ^{137}Cs vary greatly, depending on the actual Chernobyl fallout. The values for ^{137}Cs activity concentration in Table 4 were measured over the last 15 years. According to Betsou et al. (48) and Cevik & Celik (49), the ecological half-life of ^{137}Cs in moss is between 2.1 and 22 years, depending on the species. Our findings indicate that the investigated area was not a hot spot during the Chernobyl fallout.

Compared to other reports (Table 4), our measurements show no indication that ^{40}K present in moss is the result of anything else but K present in in all living organisms.

Normally, ^{232}Th, ^{238}U, and ^{226}Ra are not present in the air in Croatia (45) in measurable quantities, which points to a local source – more specifically dust – in a similar fashion as described for Pb in moss in a park (42) – and explains high values, among the highest reported (53). As activity concentrations of ^{232}Th and ^{238}U are normally independent of each other, the weak correlation (correlation coefficient of 0.78) we found suggests the same origin. While ^{226}Ra and ^{238}U are part of the same decay chain, their chemical properties are different enough that their concentrations routinely

Table 3 Comparison of average activity concentration between measurement groups

Group	Activity concentration (Bq/kg)					
	^{40}K	^{137}Cs	^{232}Th	^{238}U	^{226}Ra	^{210}Pb
1	278	13.4	19.4	27.4	13.6	483
2	263	11.8	11.8	17.8	14.5	567
3	214	17.3	12.9	19.4	16.8	605
4	211	30.4	7.17	<DL	2.26	437

DL – detection limit, the lowest quantifiable value

differ both in soil and in living organisms (58–60). Therefore, it is no surprise that the correlation between ^{226}Ra and ^{238}U values was marginal (correlation coefficient of 0.54).

Measured ^{210}Pb activities are high in view of recent literature, even higher than around some coal-fired power plants (56, 57), but not as high as reported for some locations in Bosnia and Herzegovina (55) and Greece (47). ^{210}Pb in the atmosphere is the result of the leakage of ^{222}Rn from soil and ^{210}Pb indicates the presence of ^{222}Rn in the air. Without a detailed long-term study of local weather patterns, we cannot be positive if the ^{210}Pb detected in moss is the result of local ^{222}Rn release or of ^{222}Rn transport from afar. While ^{222}Rn is considered dangerous to humans, the danger is real in closed spaces, where it can accumulate in the air. Out in the open, ^{222}Rn is not considered dangerous, especially not for short exposure times (hours or days) (61).

CONCLUSION

Our measurements of $H^*(10)$ along the Brenzberg-Točak and Žervanjska quarry fields and at two locations by Lake Orahovac show that the Radlovac Quarry complex does not present a radiological threat to Park visitors and environment, as they kept within the typical range of values of background $H^*(10)$ in Croatia.

Activity concentrations of naturally occurring radionuclides ^{232}Th, ^{238}U, ^{226}Ra, ^{7}Be, ^{210}Pb, and ^{40}K and of anthropogenic isotope ^{137}Cs in moss support these findings and do not stand out in respect to activity concentrations in moss reported in the rest of Croatia or neighbouring countries.

Conflict of interests

None to declare.

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Table 4 Comparison of our results with other studies

Area	Activity concentration (Bq/kg)					
	^{137}Cs	^{40}K	^{232}Th	^{238}U	^{226}Ra	^{210}Pb
Plitvice, Croatia (40)	14.7–510.9	84.9–194	2.03–8.92	<DL–12.3	5.9–23.6	67.9–369
Beograd area, Serbia (46)	9–221	110–490	<DL–45	<DL–80	<DL–75	
Northern Greece (47)	0–425	120–750				147–1920
Salzburg area, Austria (48)	1145–14092	51–319				
Odur province, Turkey (49)	31–469	350–1440				
Bosnia and Herzegovina (50)	4–1612					
Eastern Serbia (39)		64–484	1.4–28	1.1–50	1.1–41	
Eastern Serbia (51)		25–427	1.0–37	0.4–28	0.3–36	
Palong area, Malaysia (52)			2.8–14.0	1.44–7.68		
Serbia (53)			<DL–17		2.2–36	526–881
Thailand (53)			2.5–327		<DL–300	199–660
Sobieswo island, Poland (54)				1.36–3.87		133–501
Bosnia and Herzegovina (55)						<DL–2000
Thrace region, Turkey (41)						178–852
Western Turkey (56)						200–650
North-western Turkey (57)						219–724
Papuk, Croatia	0.53–35.0	150–437	5.92–29.2	<DL–39.1	<DL–37.3	233–996

DL – detection limit, the lowest quantifiable value

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Radiološki utjecaj aktivnoga kamenoloma u Parku prirode Papuk

Za razliku od većine sličnih parkova i rezervata prirode u svijetu, u Parku prirode Papuk nalaze se aktivni kamenolomi. U kamenolomima se izvlači kamen iz tla, pri čemu se stvara prašina te se otkrivaju dublji, potencijalno radioaktivniji slojevi. Budući da šumski putovi u Parku prirode Papuk vode do samih kamenoloma, važno je odrediti njihov radiološki učinak na taj park prirode. Mjerili smo ekvivalent ambijentalne brzine doze $H^*(10)$ i uzorkovali smo mahovinu na 26 lokacija oko dvaju od četiriju kamenoloma, oko ceste između njih te uz Orahovačko jezero, popularno turističko odredište u blizini kamenoloma. Mahovina je standardni bioindikator za teške metale, uključujući i radionuklide. Gamaspektrometrijskom smo metodom u uzorkovanim mahovinama odredili koncentracije aktivnosti ^{137}Cs i reprezentativnih prirodnih radionuklida: ^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th, i ^{40}K. Na odabranim je lokacijama $H^*(10)$ sličan onomu koji se kontinuirano mjeri drugdje u Republici Hrvatskoj. Rasponi izmjerenih vrijednosti koncentracija aktivnosti ^{137}Cs i odabranih prirodnih radionuklida u mahovinama značajno se ne razlikuju od raspona izmjerenih vrijednosti koncentracija aktivnosti navedenih radionuklida u mahovinama uzorkovanima drugdje u Hrvatskoj i u susjednim zemljama.

KLJUČNE RIJEČI: ^{137}Cs; ekvivalent ambijentalne brzine doze $H^*(10)$; gamaspektrometrija; mahovina; prirodni radionuklidi