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Original scientific paper

Radioactivity of sand from several renowned public beaches and assessment of the corresponding environmental risks

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Abstract: The radiological risk due to the presence of natural and man-made radionuclides in beach sands from several renowned seaside and riverbank public beaches was estimated in this study. The exposure levels to terrestrial radiation of the beaches were determined, as well as hazards due to human use of the analyzed sands in industry and in building constructions. Specific radionuclides concentrations in the sand samples were determined by standard gamma-spectrometry. The corresponding radiation hazards arising due to the use of sand as a building material were estimated by three different radiological hazard indices. The total absorbed gamma dose rate in the air was determined and the corresponding annual effective dose outdoors was estimated. The obtained data are relevant both from human health and environmental monitoring aspects.

Keywords: environmental radioactivity; sand; radiological hazard indices; gamma spectrometry; dose rates.

INTRODUCTION

Natural radioactivity is present in the human environment due to the presence of cosmogenic and primordial radionuclides in the Earth's crust. Cosmogenic radionuclides are produced by the interaction of cosmic-rays with atomic nuclei in the atmosphere, while primordial ones (terrestrial background radiation) were formed by the process of nucleo-synthesis. Only those radionuclides with half-lives comparable to the age of the Earth, *e.g.*, ^{40}K and members of the uranium and thorium series, can still be found today in different geological materials. Gamma radiation from these radionuclides represents the main external source of irradiation of the human body and can be considered as the largest contributor to the external dose absorbed by the population of the world.[1,2]

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Specific levels of terrestrial background radiation mainly depend on the geological and geographical conditions. Higher levels are usually associated with igneous rocks. Weathering and erosion of both igneous and metamorphic rocks in the environment transform rocks into sand deposits, some constituent minerals of which bear natural radionuclides from the uranium and thorium series as well as potassium. The study of the concentrations of radionuclides and their distribution in sands enables the assessment of radiological risk due to external human exposure to gamma radiation outdoors and inhalation of airborne radioactivity emanating from building constructions and dwellings.[3]

The objective of this was to identify and quantify significant gamma-emitting radionuclides in sand samples from several renowned world beaches, regardless their geo-genesis or chemical composition. For this purpose, sand samples were analyzed by high-resolution gamma-radiation spectrometry and the specific activities of ^{226}Ra, ^{232}Th and ^{40}K and ^{137}Cs were determined. In addition, energy-dispersive X-ray fluorescence spectrometry (EDXRFS) was employed for the semi-quantitative elemental analysis of the sands.

Based on radioactivity data, the radiation hazards due to the presence of specified radionuclides in sands commonly used in building constructions were assessed by the following three indices: the radium equivalent activity, Ra_{eq}, the representative level index I_r and the external hazard index H_{ex}.[4–6] Additionally, the absorbed dose rate in air (outdoors) due to the uniform distribution of isotopes from the uranium and thorium series and ^{40}K in the beach soil 1 m above the ground surface was estimated, and the corresponding annual effective dose (mSv yr^{-1}) outdoors calculated as the measure of human exposure to radiation.

EXPERIMENTAL

Sampling and sample preparation

Superficial beach sand samples were collected from the sea coastal sites: Ulcinj City Beach (Ulcinj, Montenegro), Great Beach of Ulcinj (Ulcinj, Montenegro), Patara (Xanthos, Turkey), Tayura (Tripoli, Libya), Tariq-City (Tripoli, Libya), Al Masif Albalady (Tripoli, Libya), Gargaresh (Tripoli, Libya), Qarit-City (Tripoli, Libya), Janzour (Tripoli, Libya), Manhattan Beach (Los Angeles, USA), Santa Monica City Beach (Santa Monica, USA), Great Salt Desert (Salt Lake City, USA) and Copacabana (Rio de Janeiro, Brazil). Some samples were collected from the Lido River Beach (Belgrade, Serbia), located on the sediment Great War Island (Danube River) in Belgrade.

The Great Beach of Ulcinj, Montenegro is located in the southern part of the eastern Adriatic Coast, 5 km from the city of Ulcinj. It is 13 km long and about 60 m wide, having an estimated capacity of about 15000 people. It is completely covered with finest grey sand.

The Patara Beach near Xanthos, Turkey is a 14.2 km long beach, located on the Mediterranean coast in southern Turkey, covered with fine brown sand and backed by swooping dunes.

All the Tripoli, Libya beaches mentioned in this work are parts of the very long Libyan Coast on the Mediterranean See, with sand of similar characteristics.

The Manhattan Beach and the Santa Monica City Beach are two parts of a much longer sandy beach in the Santa Monica Bay. The former is 3.4 km long, while the latter is 5.6 km. Both are located in the Los Angeles area (southern California).

The Copacabana, Brazil is probably the world's most famous and most popular beach. It is four kilometers long and located in the heart of Rio de Janeiro, covered with fine light yellow sand.

The Lido, Belgrade is a beach on the Great War Island, which is a river island, located in the heart of the city of Belgrade (Serbia), at the confluence of the Sava and Danube Rivers. It is covered with fine light gray sand, which can also be found on some other places along the Danube River in this area.

Approximately 2 kg of sand was taken from 50×50 cm^2 surface areas (0–10 cm depth) at each sampling site. Samples from the same location were taken at points distant from each other, about 1.5 to 2 km along the beaches. Samples from all the beaches were morphologically similar with 0.2–1.0 mm diameter grains. After the bulk samples had been homogenized and dried at 110 °C in an oven, about 450 cm^3 was transferred into cylindrical Marinelli beakers, weighed and sealed. After 40 days, the secular radioactive equilibrium between ^{226}Ra, ^{232}Th and their daughter products was attained and the samples were ready for gamma spectrometric counting.

Less than 1 g of each homogenized sample was used without further treatment for non-destructive energy dispersive X-ray fluorescence spectrometric measurements.

Analyses

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the samples were determined by standard gamma spectrometry using a HP Ge detector (Canberra) with a 23 % relative efficiency and a resolution 1.8 keV for the 1332.5 keV ^{60}Co gamma line. The detector calibration was performed using a certified standard reference soil material (MIX-OMH-SZ, National Office of Measures, Budapest) spiked with ^{22}Na, ^{57}Co, ^{60}Co, ^{89}Y, ^{133}Ba and ^{137}Cs in a cylindrical Marinelli beaker geometry. The background radiation and the samples were counted for about 68000 s. The 295.21 and 351.92 keV ^{214}Pb and 1120.29 keV ^{214}Bi gamma ray lines were used to determine the ^{226}Ra activity concentration. The ^{232}Th activity concentration was determined using the 911.07 and 969.11 keV ^{228}Ac gamma lines. The activities of ^{40}K and ^{137}Cs were determined directly from the 1460.8 and 661.66 keV gamma lines, respectively. The uncertainties are given at the 90 % confidence level.

A more detailed characterization of the sands from the different locations was performed by semi-quantitative EDXRFS spectrometry. The measurements were performed using the Canberra spectrometry system with ^{109}Cd as the excitation source and a Si(Li) detector, with a detection limit of 10 ppm. Since the same measurement time of 60 ks was chosen for all samples, a comparison of the integrated areas of the peaks at wavelengths characteristic for the elements: Cl, K, Ca, Ti, Cr, Mn, Fe, Zn, Br, Rb, Sr, Y and Zr, was possible for samples originating from very different areas of the world.

Gamma irradiation hazard indices and dose rates estimation

Three different indices were calculated, *i.e.*, the radium equivalent activity, Ra_{eq}, the representative level index I_r and the external hazard index H_{ex}. In addition, the absorbed dose rate, D, in the air was estimated.

The radium equivalent activity, Ra_{eq}, defined by Beretka and Mathew (1985),[4] is the most widely used index, which can be calculated according to the equation:

$$Ra_{eq} = c_{Ra} + \frac{10}{7}c_{Th} + \frac{10}{130}c_{K} \quad (1)$$

where c_{Ra}, c_{Th} and c_{K} are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively. Here, it is assumed that the same dose rate is produced from 370 Bq kg^{-1} ^{226}Ra or 259 Bq kg^{-1} ^{232}Th or 4810 Bq kg^{-1} ^{40}K present in the same matrix.

The representative level index I_r is defined by equation:[5]

$$I_r = \frac{1}{150}c_{Ra} + \frac{1}{100}c_{Th} + \frac{1}{1500}c_{K} \quad (2)$$

This index value must be less than unity in order to keep the radiation hazard insignificant, *i.e.*, the radiation exposure due to radioactivity from construction materials is limited to 1 mSv yr-1.

The external hazard index H_{ex} is given as:[6]

$$H_{ex} = \frac{1}{370}c_{Ra} + \frac{1}{259}c_{Th} + \frac{1}{4810}c_{K} \quad (3)$$

The total absorbed dose rate, D, in the air (outdoors) due to the uniform distribution of all the ^{226}Ra and ^{232}Th series, and ^{40}K in the beach soil 1 m above the ground surface was estimated by the formula:[7]

$$D = 0.427c_{U} + 0.662c_{Th} + 0.0432c_{K} \quad (4)$$

where the constants represent conversion factors (nGy h^{-1} *per* Bq kg^{-1}) calculated by the Monte Carlo technique for radionuclides and c_U is average activity concentration of ^{238}U.

RESULTS AND DISCUSSION

Contents of natural and man-made radionuclides

The obtained values of the activity concentrations (Bq kg^{-1}) determined for the radionuclides ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in the analyzed sand samples are listed in Table I. The presented results of the contents of radionuclides in sand samples, randomly taken from both sea and river beach areas of tourist zone, show low activity concentration of ^{226}Ra and ^{232}Th, originating from the natural radioactive series, as well as naturally occurring ^{40}K. The minimum activity concentration 2.24 Bq kg^{-1} of ^{226}Ra was determined in the sand sample from the Copacabana Beach (Brazil), while the maximum value of 15.9 Bq kg^{-1} was found in the sand from the Great Beach of Ulcinj (Montenegro). The activity concentrations of ^{232}Th were in the range 2.6–17.3, with a minimum value for the Patara Beach (Turkey) and a maximum for the Manhattan Beach (USA). The differences are not so significant and are attributable to the geochemical composition and origin of rock types in a particular area.

The content of ^{40}K depended much more on the location and had the lowest value of 18.9 Bq kg^{-1} for the sand sample from the Copacabana Beach (Brazil) and relatively low values for Tripoli (Libya) and Patara (Turkey) sands. Values up to 696 Bq kg^{-1} may be noticed at the other sites, probably due to the presence of K-feldspar in the mineral matrix of the sand deposits.

TABLE I. Activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in the sand samples collected in the period from 2004 to 2007

Sampling site		c / Bq kg^{-1}			
		^{226}Ra	^{232}Th	^{40}K	^{137}Cs
Ulcinj, Montenegro	City Beach	7.4 ±1.0	9.0 ± 1.2	192 ± 18	0.43± 0.09
	Great Beach 1	15.9 ± 2.0	14.60 ± 1.9	398 ± 37	1.83 ± 0.30
	Great Beach 2	11.2 ± 1.6	17.2 ± 2.2	412 ± 38	2.16 ± 0.31
	Great Beach 3	8.9 ± 1.2	13.4 ± 1.7	338 ± 31	1.19 ± 0.20
	Great Beach 4	9.6 ± 1.2	11.8 ± 1.5	276 ± 26	2.32 ± 0.29
	Great Beach 5	10.0 ± 1.3	12.4 ± 1.6	314 ± 29	2.49 ± 0.35
	Great Beach 6	10.4 ± 1.3	12.6 ± 1.6	251 ± 24	1.99 ± 0.27
Lido, Danube River, Belgrade, Serbia	1	7.9 ± 1.0	6.41 ± 0.87	299 ± 27	1.41 ± 0.19
	2	7.6 ± 1.0	8.8 ± 1.1	307 ± 28	1.06 ± 0.17
	3	8.4 ± 1.1	9.7 ± 1.2	278 ± 25	0.59 ± 0.12
USA	Manhattan Beach, Los Angeles, CA	5.0 ± 1.1	17.3 ± 2.4	457 ± 44	0.65 ± 0.16
	City Beach, Santa Monica, CA	11.1 ± 1.8	12.5 ± 2.2	696 ± 65	1.40 ± 0.26
	Great Salt Desert, Utah	9.4 ± 1.7	11.2 ± 1.9	230 ± 25	12.8 ± 1.4
Patara Beach, Xanthos, Turkey		10.8 ± 1.2	2.56 ± 0.53	54.5 ± 6.0	<0.3
Tripoli, Libya	Tayura-City Beach	8.5 ± 1.1	4.39 ± 0.73	54.2 ± 5.9	<0.3
	Tariq-City Beach	12.2 ± 1.4	8.4 ± 1.0	82.4 ± 8.4	<0.3
	Al Masif Albalady, Tower	7.24 ± 0.89	5.91 ± 0.82	62.4 ± 6.7	<0.3
	Al Masif Albalady	14.0 ± 1.6	3.42 ± 0.62	77.6 ± 8.0	<0.3
	Gargaresh, City	7.37 ± 0.90	3.08 ± 0.51	27.5 ± 3.3	<0.3
	Qarit-City Beach	7.15 ± 0.87	3.48 ± 0.54	31.0 ± 3.6	<0.3
	Janzour	11.28 ± 1.29	3.70 ± 0.64	80.5 ± 8.2	<0.3
Copacabana, Rio de Janeiro, Brazil		2.24 ± 0.36	6.14 ± 0.75	18.9 ± 2.4	<0.3

These results are in accordance with some previous studies,[8–10] as well as world quoted values for sand minerals: 25 (7–50), 25 (10–50) and 370 (100–700) Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively.[11] The unexpectedly low natural radionuclide content found in the Brazilian sand from the Copacabana Beach suggests the presence of light minerals, such as quartz and feldspar, as the source rather than pre-Cambrian period basement rocks deposits.[12]

Main differences in the elemental composition of the analyzed sand samples are shown in Figs. 1 and 2, where the results of X-ray fluorescence spectrometry are arranged into two groups. It can be noticed that sands from the Mediterranean region (Fig. 2) have somewhat higher Ca contents, indicating the carbonate nature of the sediment rocks, while the sands from the American continent have noticeable Zr contents. The somewhat higher values of the Fe concentrations in the sand samples may have been caused by traffic-related pollution of the urban areas where the beaches are located. All other values are of the same order of

magnitude and the differences are insignificant. These results may be considered only as an indication due to the limitations of the method used and limited number of samples.

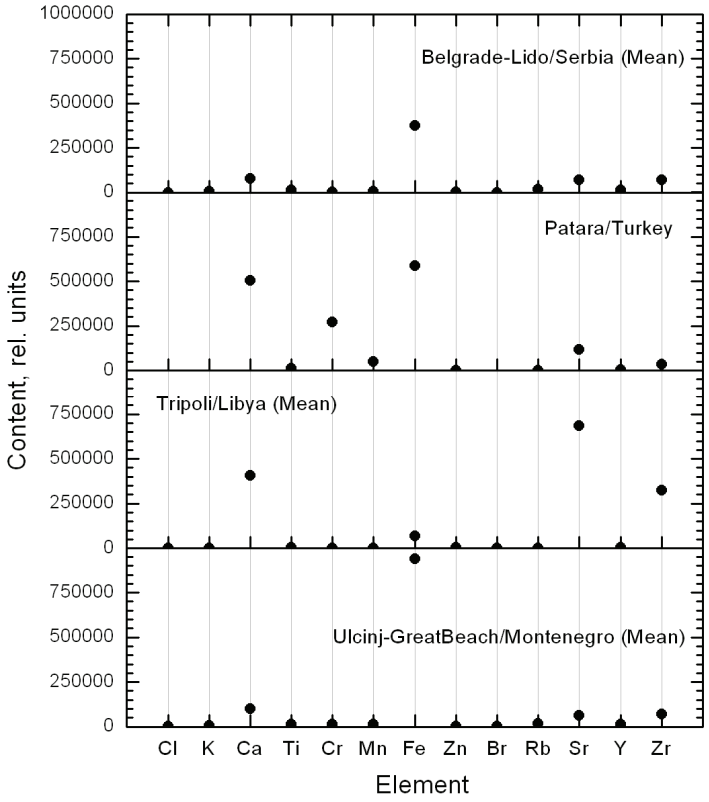

Fig. 1. Contents of some elements in the analyzed sea-and river-sand samples from the Mediterranean region.

The activity concentration of the man-made radionuclide ^{137}Cs was in the range 0.43–2.49 Bq kg^{-1} in the analyzed sand samples, with a maximum value of 12.8 Bq kg^{-1} measured in the Great Salt Desert, Utah, USA, sample radioactive cesium, a fission product with a half-life of 30.2 yr, is mostly present in the environment due to the Chernobyl nuclear accident in 1986, but it may be assumed that nuclear probes performed during sixties in this part of the world (in the neighboring state of New Mexico) also contributed. The activity concentration of ^{137}Cs becomes lower if compared with some previous results[13,14] for location sites in Serbia and Montenegro, mostly due to the selective migration and geochemical fractionation of cesium in sediments. In general, the ^{137}Cs contents were very low in all samples, especially in the Turkish and Libyan sands, where they were below the minimal detectable value.

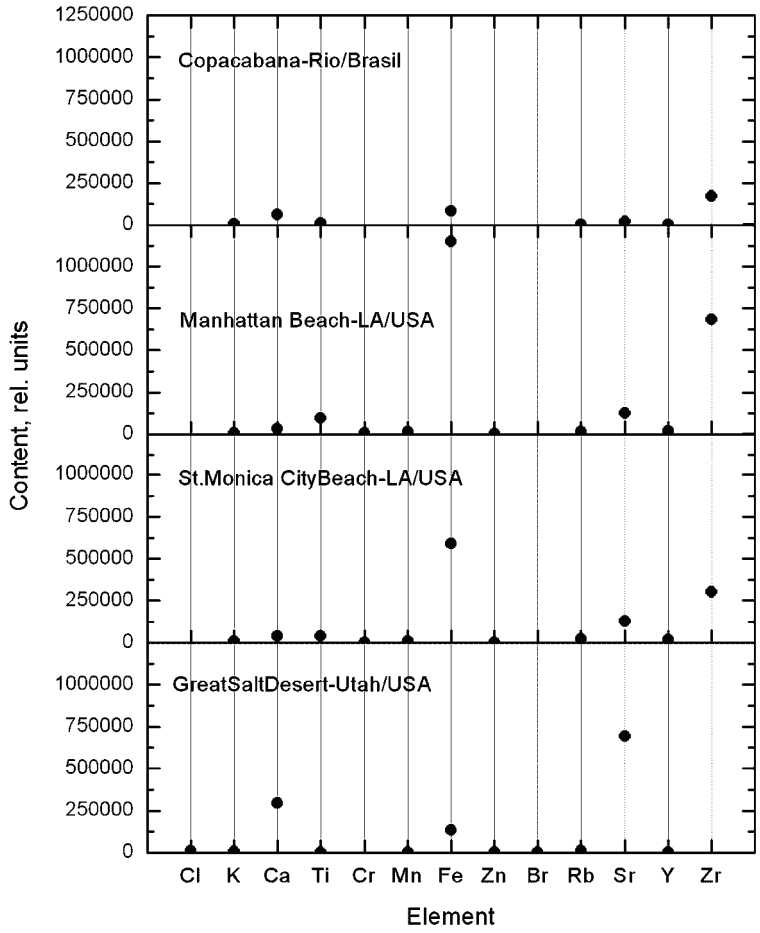

Fig. 2. Contents of some elements in the analyzed sea-sand samples from the American Continent region.

Gamma irradiation hazard indices and dose rates

As river- and sea-beach sand minerals are used in industry and in building constructions, the gamma-ray radiation hazards due to the specified radionuclides were assessed by three indices, *i.e.*, the radium equivalent activity, Ra_{eq}, the representative level index, I_r, and the external hazard index, H_{ex}. The results obtained for the sands studied in this work are presented in Table II.

The presented results show that the Ra_{eq} index for the sand samples had values in the range 12.5 (Rio de Janeiro, Brazil) to 82.5 Bq kg^{-1} (Santa Monica, CA, USA), compared with the population-weighted average value of global primordial radiation of 59 nGy h^{-1}.[2] This index is related to the external gamma dose and internal dose due to radon and its daughter products and allows a comparison of the activities and radiological effects of sediment samples containing

different concentrations of radionuclides. The calculated values of I_r were within 9.0–66 % of the 1 Bq kg^{-1} limit value.

TABLE II. Gamma radiation hazard indices for the analyzed sands: radium equivalent activity, Ra_{eq}, representative level index, I_r, external hazard index, H_{ex}, and the corresponding absorbed dose, D, and annual effective dose, E

Sampling site		Ra_{eq} Bq kg^{-1}	I_r Bq kg^{-1}	H_{ex} Bq kg^{-1}	D nGy h^{-1}	E mSv yr^{-1}
Ulcinj, Montenegro	City Beach	35.1	0.27	0.18	17.4	0.041
	Great Beach 1	67.4	0.52	0.09	33.7	0.021
	Great Beach 2	67.5	0.52	0.18	33.9	0.041
	Great Beach 3	54.1	0.42	0.15	27.3	0.033
	Great Beach 4	47.7	0.37	0.13	23.8	0.029
	Great Beach 5	51.9	0.40	0.14	26.1	0.032
	Great Beach 6	47.7	0.36	0.13	23.6	0.029
Lido, Danube River, Belgrade, Serbia	1	40.0	0.32	0.11	20.6	0.025
	2	43.7	0.34	0.12	22.3	0.027
	3	43.7	0.34	0.12	22.0	0.027
USA	Manhattan Beach, Los Angeles, CA	64.9	0.51	0.18	33.4	0.041
	City Beach, Santa Monica, CA	82.5	0.66	0.22	43.1	0.053
	Great Salt Desert, Utah	43.2	0.33	0.12	21.4	0.026
Patara Beach, Xanthos, Turkey		18.7	0.13	0.05	8.7	0.011
Tripoli, Libya	Tayura-City Beach	19.0	0.14	0.05	8.9	0.011
	Tariq-City Beach	30.5	0.22	0.08	14.3	0.018
	Al Masif Albalady, Tower	20.5	0.15	0.06	9.7	0.012
	Al Masif Albalady	24.9	0.18	0.07	11.6	0.014
	Gargaresh, City	13.9	0.10	0.04	6.4	0.008
	Qarit-City Beach	14.5	0.10	0.04	6.7	0.008
	Janzour	22.8	0.17	0.06	10.7	0.013
Copacabana, Rio de Janeiro, Brazil		12.5	0.09	0.03	5.8	0.007

All gamma-ray absorbed dose rates, D, presented in Table II are within the range 5.8–43.1 nGy h^{-1}, *i.e.*, less than the world average of 55 nGy h^{-1}.[11] Finally, in order to obtain a rough estimate for the annual effective dose outdoors, the conversion coefficient from the absorbed dose in air to the effective dose and the outdoors occupancy factor had to be taken into account. As in the UNSCEAR reports (1993, 2000), a conversion coefficient of 0.7 Sv Gy^{-1} from the absorbed dose in air to the effective dose received by adults and an outdoor occupancy factor of 0.2 were used. The annual effective dose E (mSv yr^{-1}) outdoors was then calculated using the following formula:

$$E = D \times 24\ \text{h} \times 365.25\ \text{d} \times 0.2 \times 0.7 \times 10^{-3} \quad (5)$$

The obtained *E* values for all the analyzed sands were lower than the world-wide outdoors annual effective dose average of 0.07 mSv yr^{-1},[2] and also below the value of 1.0 mSv yr^{-1}, recommended by the International Commission on Radiological Protection[15] as the maximum allowed annual dose for the public.

CONCLUSIONS

This study showed that the analyzed sand samples from different world beaches had various radionuclide contents within the average world quoted values. The corresponding gamma radiation hazard indices and annual effective dose were below those of the limits considered acceptable.

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ИЗВОД

РАДИОАКТИВНОСТ ПЕСКА СА НЕКОЛИКО ПОЗНАТИХ ЈАВНИХ ПЛАЖА И ПРОЦЕНА ОДГОВАРАЈУЋИХ РИЗИКА ПО ЖИВОТНУ СРЕДИНУ

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У раду су приказани резултати процене радиолошке опасности која је последица присуства природних и произведених радионуклида у песку са неколико познатих морских и речних јавних плажа. Одређени су нивои изложености људи зрачењу из песка на плажама као и опасност услед коришћења анализираних песака у индустрији и грађевинарству. Концентрације значајнијих радионуклида у узорцима су одређене стандардном гама-спектрометријом. Радиолошка опасност услед коришћења песка као грађевинског материјала процењена је на основу три индикатора радиолошког ризика. Одређена је укупна апсорбована доза гама зрачења у ваздуху и процењене су одговарајуће годишње ефективне дозе услед боравка на плажама. Добијени резултати су битни са аспекта заштите људског здравља као и мониторинга животне средине.

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