GAMMA SPECTROMETRY ANALYSIS OF NATURAL AND MAN-MADE RADIOACTIVITY AND ASSESSMENT OF RADIOLOGICAL RISK IN SOILS AROUND STEEL INDUSTRY

ANTOANETA ENE[1*], ANA PANTELICĂ[2], FLORIN SLOATĂ[1], HESHAM M.H. ZAKALY[3], HUSEYIN OZAN TEKIN[4]

[1] Dunarea de Jos University of Galati, Faculty of Sciences and Environment, Department of Chemistry, Physics and Environment, INPOLDE Center, 47 Domneasca St., 800008 Galati, Romania
[2] Horia Hulubei National Institute for R&D in Physics and Nuclear Engineering, 30 Reactorului St., 077125 Magurele, Ilfov County, Romania
[3] Institute of Physics and Technology, Ural Federal University, Yekaterinburg 620002, Russia
[4] University of Sharjah, College of Health Sciences, Medical Diagnostic Imaging Department, Sharjah 27272, United Arab Emirates
[*] *Corresponding author*: aene@ugal.ro

Received April 7, 2023

Abstract. In this paper low background high-resolution gamma spectrometry was used for the determination of the activity concentrations of the natural and man-made radionuclides in topsoil samples collected in the vicinity of a large steel plant in Galati, Romania. Their values (mean ± standard deviation) for ^{238}U, ^{226}Ra, ^{232}Th, ^{235}U, ^{210}Pb, ^{40}K and ^{137}Cs were: 38.1 ± 6.2; 35.9 ± 5.4; 35.5 ± 3.0; 1.8 ± 0.4; 47.3 ± 8.6; 504.5 ± 38.9 and 7.2 ± 4.7 Bq·kg^{-1}, respectively. ^{60}Co and ^{241}Am had activities below the detection limit: 0.6–1.3 and 2.2–4.7 Bq·kg^{-1}, respectively. The results indicate inputs of natural radionuclides in the sites close to the slag dump, raw materials transportation facility, ore and coal processing factories, blast furnaces, and iron scrap deposit. The evaluation of the associated radiological impact was performed based on selected hazard parameters: absorbed gamma dose rate, total external gamma dose rate, radium equivalent activity index, external hazard index, representative level index, annual effective dose, excess lifetime cancer risk, and annual gonadal dose.

Key words: radioactivity, soils, risk, gamma spectrometry, steel industry, Galati, Romania.

DOI: https://doi.org/10.59277/RomJPhys.2023.68.803

1. INTRODUCTION

Humans have been exposed to ionizing radiation in environment from natural and anthropogenic radiation sources [1, 2] which impacts different body organs and tissues in various proportions [2–4]. Natural radiation is due to the terrestrial (primordial) radionuclides existing in the Earth's crust (uranium (^{238}U), actinium (^{235}U) and thorium (^{232}Th) radioactive series and the nonseries natural radionuclide ^{40}K) [5–12] and the cosmogenic radioisotopes produced in the outer space [13–16].

The artificial radioactivity is due to the fallout from accidents which took place in nuclear reactors (*e.g.* Chernobyl accident) and nuclear weapons testing, as well as the cycle of the nuclear fuel in nuclear installations [1, 2, 14, 17]. Terrestrial gamma emitters have different concentrations in soils around the world, in relation with the geological background of each region, landscape features, soil lithology, mineralogy and physico-chemical characteristics [5, 18–22] and determine the main part of the external exposure of people to radiations [1, 2]. Many industrial practices which involve naturally occurring radioactive materials (NORMs) (mining, oil extraction, phosphate fertilizers production, metal production, processing of minerals, iron ores and coal) and transportation and disposal of industrial wastes and radionuclides-bearing materials, may increase the risk of soil contamination with radioactive isotopes in the vicinity of industrial installations.

The monitoring of environmental gamma radiation for human protection requires the development of performant equipment capable of carrying out accurate measurements. Gamma-ray spectrometry proved to be a powerful non-destructive analytical tool for the qualitative and quantitative determination of the gamma emitters in a variety of materials with different matrices, such as soils, sediments, rocks, phosphates, building materials, plants, etc. [6, 11, 12, 20, 23, 24]. Over the past years, technical and procedural improvements in low background gamma-spectroscopic analysis of NORMs and artificial isotopes were developed to obtain better detection limits of radionuclides in environmental compartments [25] and industrial byproducts and wastes [23, 25].

Due to emissions specific to ironmaking and steelmaking at Galati, SE Romania [23, 26, 27], the soils around the metallurgical industrial area exhibit a high level of contamination with metals and organic pollutants [28–31] which could impact the agroecosystems of Danube River region [32]. NORMs could be transported from Danube ore harbour to the steel enterprise (Fig. 1) and the dust particles might trap the radionuclides and be transported over large areas. Thus, the determination of the distribution of the natural and anthropogenic radionuclides in soils around facilities of iron and steel industry is of outermost importance for the assessment of public exposure and building a national database with radioactivity levels due to industrial activities.

To the best of our knowledge, data for soil and sediment radioactivity in Romania are very scarce [12, 25, 33–36] and information about radionuclides in soils around metallurgical industry, the associated dose and natural radioactivity risk to population lacks in the literature.

The aim of this paper was to investigate the activity concentration of the main natural and man-made radionuclides in topsoil neighboring the integrated steel plant at Galati, Romania by low background high resolution gamma spectrometry technique, and to assess the radiological and health risk of gamma-emitting radionuclides present in soil, as well as population exposure dose in the region.

2. MATERIAL AND METHODS

2.1. SAMPLE COLLECTION AND PREPARATION

Soil samples were collected from the surface layer (0–5 cm) in the vicinity of the iron and steel industrial enterprise located in the western part of Galati municipality, Lower Danube basin, SE part of Romania (Fig. 1). The GPS coordinates and characteristics of the sampling sites are presented in Table 1. The enterprise is linked in the SE part with the mineral (ore) harbour on the Danube River through an ores' conveyor belt. The agglomeration/sintering plants and coke factory are in the S part, the blast furnaces in SW, the slag dump and internal transportation infrastructure in W and the iron scrap deposit and lime factory in N. Composite samples (1–2 kg) were packed in labeled polyethylene bags and brought to the laboratory where were cleaned for pebbles, vegetation remains and other debris.

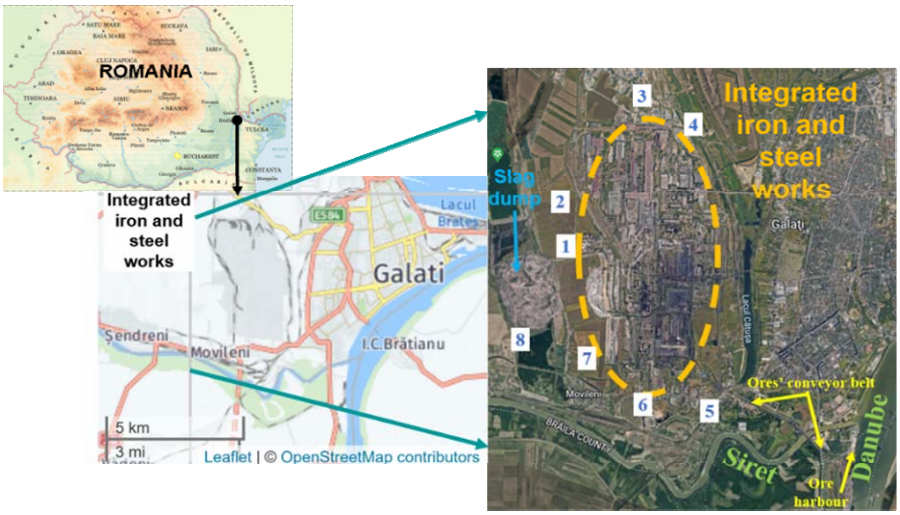


Fig. 1 – Schematic map of sampling sites around the integrated steel works at Galati, Romania.

Table 1

Characteristics of soil sampling sites from Fig. 1

Site no.	Latitude	Longitude	Altitude (m)	Location description
1	45.431403	27.957054	49	Access route of slag dump
2	45.439593	27.948065	43	N of slag dump
3	45.459372	27.971731	56	N of enterprise, Atlas area
4	45.452881	27.984224	61	North Gate of enterprise
5	45.405227	27.969345	37	SE of enterprise, Tirighina area
6	45.408567	27.975417	39	South Gate of enterprise
7	45.411653	27.960171	38	SW of enterprise, Movileni area
8	45.416060	27.944476	9	S of slag dump, Movileni area

The samples were prepared for gamma-spectrometric analysis at Laboratory of Experimental Nuclear Physics and Dosimetry, INPOLDE research center, Dunarea de Jos University of Galati, Romania, being dried at 110°C up to 48 h, crushed, ground, homogenized, and passed through a 2 mm mesh size sieve.

2.2. GAMMA RAY SPECTROMETRY

The low-background high resolution gamma-ray spectrometry technique was applied at GamaSpec laboratory of Horia Hulubei National Institute of Physics and Nuclear Engineering (IFIN-HH) in Magurele, Romania, to determine the natural and artificial radioactivity of industrial topsoils.

The gamma spectrometric chain was equipped with a coaxial GEM-30185 Ortec HPGe detector of 2.3 keV energy resolution at 1332 keV of ^{60}Co and 30% relative efficiency, placed in a lead shield of 10 cm thickness, coated inside with foils of 1 mm Sn and 1.5 mm Cu thickness to reduce the ambient background radiation in the spectra. The background count rate in the energy range from 20 to 2700 keV was lower than 1.9 cps (counts per second) for a counting time of 24 h. GAMMAW software program was used for the spectra processing [23].

The soil samples (around 100 g each) were measured in cylindrical polyethylene beakers (7.2 cm diameter and 2.5 cm height) placed on the detector end cap, for counting times of 5.3 to 20.5 h, after more than 3 weeks of keeping sealed in the measuring box to establish the radioactive equilibrium between ^{226}Ra and its gaseous radioactive descendant ^{222}Rn (radon) of 3.8 d half-life in the ^{238}U series.

2.3. RADIOLOGICAL RISK PARAMETERS

To assess the radiological and human health risk of soil radioactivity, we computed the dose rates due to terrestrial (natural and artificial) radionuclides, the total dose rate (taking also into account the cosmic component), the annual effective dose and the associated radiological parameters, synthesized and adapted after various publications [3, 4, 7, 10, 37–43], the complete proposed model being described below.

The outdoor absorbed gamma dose rate in air at 1 m above ground due to natural radionuclides in soil, DR_{NAT} (nGy·h^{-1}), was calculated from the concentration activities c(Ra), c(Th) and c(K) (in Bq·kg^{-1}) of ^{226}Ra, ^{232}Th and ^{40}K, respectively, as:

$$DR_{NAT}\ (nGy \cdot h^{-1}) = 0.462\ c(Ra) + 0.604\ c(Th) + 0.0417\ c(K) \quad (1)$$

where the values of 0.462, 0.604 and 0.0417 are the activity to-outdoor dose conversion factors for the natural radionuclides, expressed in nGy·h^{-1} per Bq·kg^{-1}.

The gamma dose rate due to artificial radionuclide ^{137}Cs existent in soil in a concentration c(Cs), in Bq·kg^{-1}, was estimated by using in the equation (2) the dose conversion coefficient of 0.1243 nGy·h^{-1} per Bq·kg^{-1}, proposed by several authors [17, 42], value close to that obtained by Clouvas *et al.* (2000) [44] using the Monte Carlo simulation (GEANT code of CERN):

$$DR_{Cs}\ (nGy \cdot h^{-1}) = 0.1243\ c(Cs) \quad (2)$$

resulting a terrestrial gamma dose rate DR_{TER}:

$$DR_{TER} = DR_{NAT} + DR_{Cs} \quad (3)$$

The total external gamma dose rate, DR_{TOT}, is due to both terrestrial (DR_{TER}) and cosmic (DR_{COS}) component contribution of the ionizing radiation:

$$DR_{TOT} = DR_{TER} + DR_{COS} \quad (4)$$

where the estimate of the cosmic ray dose rate $DR_{COS}(z)$ at elevation z (in km) above the sea level is given by the formula:

$$DR_{COS}(z) = DR_{COS}(0)\ (0.21\ e^{-1.649\,z} + 0.79\ e^{0.4528\,z}) \quad (5)$$

assuming a dose rate $DR_{COS}(0) = 32\ nGy \cdot h^{-1}$ at the sea level in the northern hemisphere, for latitudes considered here (from 40°N to 50°N).

With the notations given above, the radiological parameters − radium equivalent activity index (Ra_{eq}), external hazard index (H_{ex}), representative level index (I_G), annual effective dose (AED), and excess lifetime cancer risk (ELCR) − have been assessed using the following equations:

$$Ra_{eq}\ (Bq \cdot kg^{-1}) = c(Ra) + 1.43\ c(Th) + 0.077\ c(K) \quad (6)$$

$$H_{ex} = c(Ra)/(370\ Bq \cdot kg^{-1}) + c(Th)/(259\ Bq \cdot kg^{-1}) + c(K)/(4810\ Bq \cdot kg^{-1}) \quad (7)$$

$$I_G = c(Ra)/(150\ Bq \cdot kg^{-1}) + c(Th)/(100\ Bq \cdot kg^{-1}) + c(K)/(1500\ Bq \cdot kg^{-1}) \quad (8)$$

$$AED\ (mSv \cdot y^{-1}) = DR_{TER}\ (nGy \cdot h^{-1}) \cdot T \cdot OF \cdot DCF \cdot 10^{-6} \quad (9)$$

$$ELCR = AED\ (mSv \cdot y^{-1}) \cdot LS \cdot PC \cdot 10^{-3} \quad (10)$$

where $T = 8760\ h \cdot y^{-1}$ is the number of hours per year, $OF = 0.2$ is the outdoor occupancy factor, $DCF = 0.7\ Sv \cdot Gy^{-1}$ is the dose conversion factor, LS is the lifespan (70 years), PC is the probability coefficient for detriment-adjusted cancer risk ($0.055\ Sv^{-1}$ for stochastic effects that radiation may have on public health [37]).

In the case of utilization of the soil in the dwellings construction, an important reproductive organs safety parameter, used for alerting the local people to avoid its utilization, is the annual gonadal dose equivalent (AGD) [3, 39, 45], which relates the impact of ionizing radiation to the most sensitive parts of the human body:

$$AGD\ (mSv \cdot y^{-1}) = [3.09\ c(Ra) + 4.18\ c(Th) + 0.314\ c(K)] \cdot 10^{-3} \quad (11)$$

The conversion coefficients in equation (11) are expressed in $Sv \cdot y^{-1}$ per $Bq \cdot kg^{-1}$.

3. RESULTS AND DISCUSSION

3.1. ACTIVITY OF NATURAL AND ARTIFICIAL RADIONUCLIDES IN INDUSTRIAL SOIL

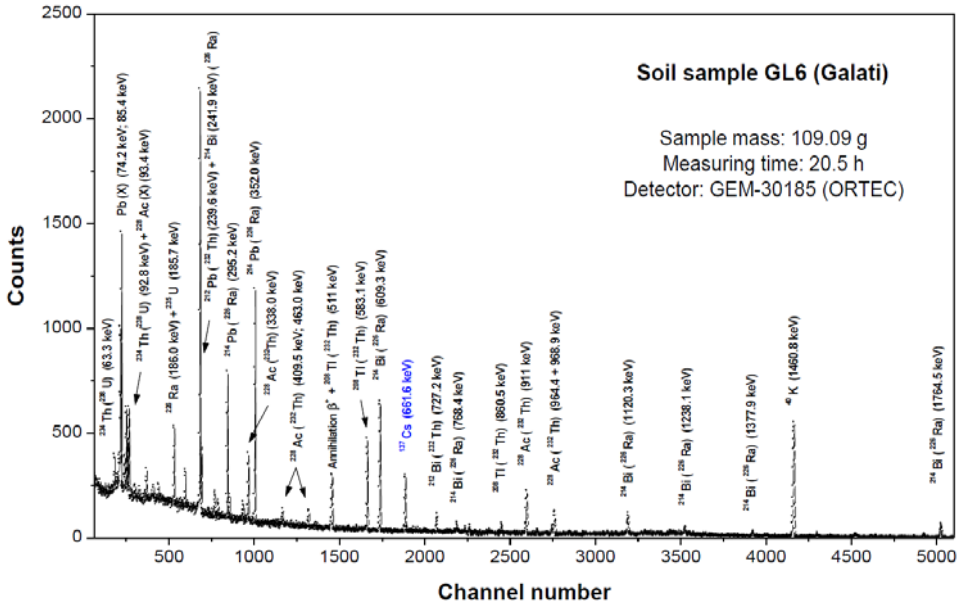
The analyzed natural radionuclides were the following: ^{226}Ra (from ^{214}Pb and ^{214}Bi activities, progenies of ^{222}Rn from ^{238}U–^{226}Ra series), ^{238}U (from ^{234}Th activity), ^{232}Th (from ^{228}Ac, ^{212}Pb and ^{208}Tl activity), ^{235}U (^{226}Ra spectral interference correction for 185.7 keV), ^{210}Pb (^{238}U–^{226}Ra series) and ^{40}K. The investigated artificial radionuclides were ^{60}Co, ^{137}Cs and ^{241}Am, but the concentrations of ^{60}Co and ^{241}Am were below detection limits. The activity concentrations of all radionuclides are given in Table 2, together with analytical uncertainties (coverage factor $k = 1$) due to counting statistics, detection efficiency, and gamma-ray emission intensity uncertainties. Also, the concentrations of U, Th and K are presented in Table 2, calculated considering that 1 g natural U yields 12357 Bq ^{238}U, 1 g natural Th yields 4069 Bq ^{232}Th, and 1 g K yields 31.66 Bq ^{40}K [23]. The gamma spectra of a soil sample (Fig. 2a) and laboratory background (Fig. 2b) indicate the presence of the peaks resulted from natural radionuclides and the artificial nuclide ^{137}Cs.

Table 2

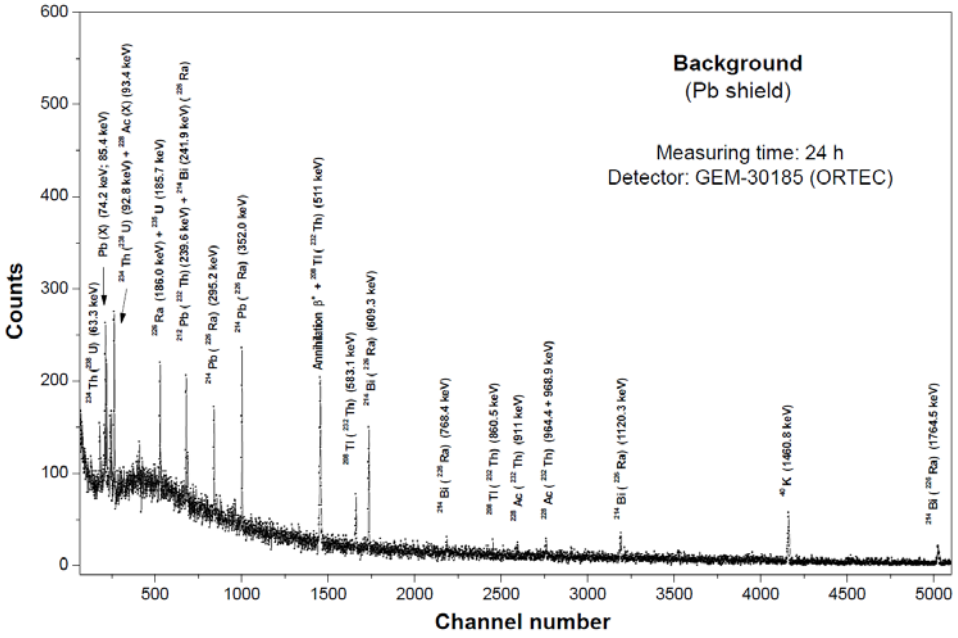
Activity concentration (Bq·kg^{-1}) of the radionuclides (RN) determined in soils collected at Galati (counting time in parentheses) and element (EL) concentration (mg·kg^{-1} for U and Th; g·kg^{-1} for K)

RN/ EL	Soil 1 (5.3 h)	Soil 2 (17.0 h)	Soil 3 (5.5 h)	Soil 4 (7.08 h)	Soil 5 (14.6 h)	Soil 6 (20.5 h)	Soil 7 (13.5 h)	Soil 8 (7.0 h)
^{226}Ra	46.3 ± 2.5	32.0 ± 1.4	36.2 ± 1.8	36.1 ± 2.1	35.4 ± 1.6	40.1 ± 1.6	32.1 ± 1.5	28.9 ± 1.7
^{214}Pb	45.1 ± 2.7	32.9 ± 2.0	36.8 ± 2.5	37.7 ± 2.9	36.1 ± 2.0	39.8 ± 2.2	32.3 ± 1.9	30.9 ± 2.1
^{214}Bi	47.5 ± 4.2	31.1 ± 1.9	35.5 ± 2.7	34.5 ± 3.2	34.7 ± 2.6	40.4 ± 2.2	31.9 ± 2.4	27.0 ± 2.8
^{238}U	47 ± 18	36 ± 11	43 ± 14	42 ± 25	28 ± 7	32 ± 7	41 ± 13	37 ± 10
^{235}U	2.1 ± 1.1	1.6 ± 0.8	2.3 ± 1.1	2.1 ± 0.8	< 2.2	< 2.1	1.2 ± 0.8	1.9 ± 1.1
^{210}Pb	43 ± 32	59 ± 20	43 ± 37	49 ± 39	57 ± 22	37 ± 31	53 ± 38	37 ± 32
^{232}Th	41.6 ± 1.9	35.0 ± 1.4	33.2 ± 1.6	35.5 ± 1.7	36.6 ± 1.4	36.7 ± 1.4	31.7 ± 1.4	33.6 ± 1.6
^{228}Ac	42.1 ± 3.3	37.0 ± 1.6	35.5 ± 2.7	36.4 ± 2.1	37.0 ± 1.9	36.6 ± 1.6	31.6 ± 2.0	33.2 ± 2.2
^{212}Pb	42.1 ± 2.8	34.1 ± 2.7	32.4 ± 2.6	35.2 ± 3.7	37.1 ± 2.4	36.8 ± 2.7	32.7 ± 2.1	33.4 ± 2.6
^{208}Tl*	40.8 ± 3.6	34.0 ± 2.8	31.8 ± 2.9	34.8 ± 2.9	35.5 ± 2.7	36.6 ± 2.7	30.6 ± 3.0	34.2 ± 3.3
^{208}Tl	14.7 ± 1.3	12.3 ± 1.0	11.5 ± 1.1	12.6 ± 1.0	12.9 ± 1.0	13.2 ± 1.0	11.1 ± 1.1	12.4 ± 1.2
^{40}K	563 ± 36	519 ± 31	435 ± 28	494 ± 30	490 ± 29	485 ± 27	508 ± 30	542 ± 37
^{137}Cs	7.3 ± 0.9	11.6 ± 1.0	13.5 ± 1.0	4.6 ± 0.7	7.9 ± 0.6	10.8 ± 0.7	1.2 ± 0.3	0.9 ± 0.4
^{241}Am	< 4.7	< 2.2	< 4.4	< 3.8	< 2.8	< 2.4	< 2.7	< 3.4
^{60}Co	< 1.3	< 0.6	< 1.2	< 1.1	< 0.8	< 0.6	< 0.7	< 1.1
U	3.8 ± 1.5	2.9 ± 0.9	3.4 ± 1.1	3.4 ± 2.0	2.2 ± 0.5	2.6 ± 0.6	3.3 ± 1.1	3.0 ± 0.8
Th	10.2 ± 0.5	8.6 ± 0.3	8.2 ± 0.4	8.7 ± 0.4	9.0 ± 0.3	9.0 ± 0.3	7.8 ± 0.3	8.3 ± 0.4
K	17.8 ± 1.1	16.4 ± 1.0	13.7 ± 0.9	15.6 ± 0.9	15.5 ± 0.9	15.3 ± 0.9	16.0 ± 0.9	17.1 ± 1.2

Note: Detection limits were determined with a probability $P = 95\%$ (2 σ); * ^{212}Bi branching decay factor of 0.3594 for ^{208}Tl was considered in ^{232}Th activity concentration calculation [23].



a)



b)

Fig. 2 – Typical gamma spectra of radionuclides from a soil sample collected from site no. 6 (laboratory code GL6) in the vicinity of Galati enterprise (a) and laboratory background (b).

The statistics summary of main radionuclides and element concentration is presented in Table 3. From Table 2 it results that highest natural radioactivity level is exhibited by the soil collected in the vicinity of the slag dump, ore/coke processing factories, blast furnaces, transportation lines and scrap deposit. The average activity concentrations (ranges in parentheses) of ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K (in Bq·kg^{-1}) (Table 3) of 35.9 (28.9–46.3), 38.1 (28.0–47.0), 35.5 (31.7–41.6), and 504.5 (435.0–563.0), respectively, were found to be slightly higher than the world and Romanian average values reported by the United Nations Scientific Committee on Effects of Atomic Radiation (UNSCEAR) [1, 2]. U, Th, and K concentration values were similar with those determined by instrumental neutron activation analysis technique in Romanian soils [28, 29] and European and world topsoil, except for K with slightly smaller values in Romania [29]. The values for ^{137}Cs activity concentration show a large variability (65.1%) and are similar with other findings [25, 35, 36].

Table 3

Descriptive statistics of radionuclides' activity and radioelement concentrations in the targeted soils

Parameter	^{226}Ra (Bq·kg^{-1})	^{238}U (Bq·kg^{-1})	^{232}Th (Bq·kg^{-1})	^{40}K (Bq·kg^{-1})	^{137}Cs (Bq·kg^{-1})	U (mg·kg^{-1})	Th (mg·kg^{-1})	K (g·kg^{-1})
Min	28.9	28.0	31.7	435.0	0.90	2.20	7.80	13.70
Max	46.3	47.0	41.6	563.0	13.5	3.80	10.20	17.80
Ave	35.9	38.1	35.5	504.5	7.23	3.09	8.73	15.93
SD	5.4	6.2	3.0	38.9	4.70	0.50	0.72	1.24
CV (%)	15.1	16.3	8.5	7.7	65.1	16.2	8.3	7.8

Ave = average; SD = standard deviation; CV = coefficient of variation.

3.2. ASSESSMENT OF RADIOLOGICAL RISK PARAMETERS

Based on the activity values and the methodology proposed in §2.3, it was calculated the outdoor absorbed dose rate at 1 m above the ground surface due to γ-rays emission in air from ^{226}Ra, ^{232}Th, and ^{40}K natural radionuclides (DR_{NAT}) and to all detected terrestrial radionuclides (DR_{TER}), including the artificial radionuclide ^{137}Cs, although this one had a small contribution to the terrestrial dose rate (Table 4). Also, Table 4 shows the results of the total absorbed dose in air, DR_{TOT}, originated both from terrestrial and cosmic radiation. The values (average/(range)) for absorbed dose rates (59.94/(55.31–70.90) nGy h^{-1} for D_{TER}, 91.97/(87.34–102.95) nGy h^{-1} for D_{TOT}) are comparable with those reported for Romania [2], 59/(20–125) and 92/(52–163) nGy h^{-1}, respectively, with a slight exceeding of the mean values in the industrial sites which recorded higher radioactivity levels.

To evaluate the radiological hazards resulting from soil radioactivity, radium equivalent activity (Ra_{eq}), representative gamma index (I_G), external hazard index (H_{ex}), annual effective dose (AED), excess lifetime cancer risk (ELCR), and annual gonadal dose rate (AGD) were calculated using Eqs. 6–11 (Table 4).

Compared with the recommended values (Ra_{eq} = 370 Bq kg^{-1}, I_G < 1, H_{ex} < 1, AED = 0.070 mSv y^{-1}, ELCR = 2.9 × 10^{-4}, AGD = 0.300 mSv y^{-1}) [3], our results indicate higher values for AED in sites nos. 1, 2, 4, 5 and 6; for I_G and ECLR in site no. 1, and for AGD in all sites. As a result, gamma radiation emitted from the soil samples around Galati steel industry might pose health hazards for the inhabitants of the area.

Table 4

Radiological indices in the investigated soil samples and descriptive statistics

Site no.	DR_{NAT} (nGy·h^{-1})	DR_{TER} (nGy·h^{-1})	DR_{TOT} (nGy·h^{-1})	Ra_{eq} (Bq·kg^{-1})	I_G	H_{ex}	AED (mSv·y^{-1})	ELCR (×10^{-4})	AGD (mSv·y^{-1})
1	69.99	70.90	102.95	149.1	1.10	0.40	0.087	3.0	0.494
2	57.57	59.01	91.04	122.0	0.91	0.33	0.072	2.5	0.408
3	54.92	56.59	88.65	117.2	0.86	0.32	0.069	2.4	0.387
4	58.72	59.29	91.36	124.9	0.93	0.34	0.073	2.5	0.415
5	58.86	59.85	91.87	125.4	0.93	0.34	0.073	2.6	0.416
6	60.85	62.19	94.22	129.8	0.96	0.35	0.076	2.7	0.429
7	55.16	55.31	87.34	116.5	0.87	0.31	0.068	2.4	0.391
8	56.25	56.36	88.36	118.7	0.89	0.32	0.069	2.4	0.400
Min	54.92	55.31	87.34	116.5	0.86	0.31	0.068	2.4	0.387
Max	69.99	70.90	102.95	149.1	1.10	0.40	0.087	3.0	0.494
Ave	59.04	59.94	91.97	125.5	0.93	0.34	0.074	2.6	0.418
SD	4.86	4.96	4.96	10.6	0.08	0.03	0.006	0.2	0.034
CV (%)	8.2	8.3	5.4	8.4	8.1	8.4	8.3	8.3	8.1

4. CONCLUSIONS

Terrestrial gamma emitters have different concentrations around the world, in relation with the geographical position and base rock formations, and their activity concentrations determination is very important for the evaluation of external exposure of people to ionizing radiation. Low background high resolution gamma-spectrometry was employed to detect and quantitatively analyze the natural and artificial radionuclides in industrial soils.

Radiological risk was assessed using the parameters: radium equivalent activity, representative gamma index, external hazard index, annual effective dose, excess lifetime cancer risk, and annual gonadal dose rate. Moreover, the total absorbed dose rates were evaluated as the sum of terrestrial and cosmic radiation contribution. The study fills a gap in the database of natural radionuclide levels in industrial soils and related risk parameters for human health in the vicinity of integrated steel plants. Also, information obtained for the radiocesium activity in soils in SE Romania is valuable for the mapping of this fallout radionuclide in this region of the world and tracking the changes in its concentration.

Acknowledgments. This work was supported by the research grant with contract no. 9187/2023, funded by Dunarea de Jos University of Galati. The first author (AE) would like to thank Professor Alexandros Clouvas from Aristotle University of Thessaloniki, Greece, for useful discussions.

REFERENCES

1. United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), *Report to the General Assembly, with scientific annexes*, Vol. 1, Sources and Effects of Ionising Radiation, United Nations, New York, 2000.
2. United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), *Report of the United Nations Scientific Committee on the Effect of Atomic Radiation to the General Assembly*, Vol. 1, New York, 2010.
3. H.M.H. Zakaly, M.A.M. Uosif, S.A.M. Issa, H.O. Tekin, H. Madkour, M. Tammam, A. El-Taher, G. A. Alharshan, and M. Y.A. Mostafa, Mar. Pollut. Bull. **171**, 112658 (2021).
4. D. A. E. Darwish, K. T. M. Abul-Nasr, and A. M. El-Khayatt, J. Radiat. Res. Appl. Sci. **8**, 17–25 (2015).
5. H.A. Awad, I.A.El-Leil, R.M. El-Wardany, A. Ene, A. Tolba, M. Kamel, A. Nastavkin, and H.M. Zakaly, Rom. J. Phys. **67** (3–4), 803 (2022).
6. E.S.R. Lasheen, H.M.H. Zakaly, B.M. Alotaibi, D.A. Saadawi, A. Ene, D. Fathy, H.A. Awad, and R.M. El Attar, Minerals **12**, 1385 (2022).
7. N.M. Moghazy, A.M. El-Tohamy, M.M. Fawzy, H.A. Awad, H.M.H. Zakaly, S.A.M. Issa, and A. Ene, Appl. Sci. **11**, 6471 (2021).
8. H.M.H. Zakaly, H.A. Awad, N.M. Moghazy, H.O. Tekin, A. Rabie, M.M. Fawzy, A.M. El-Tohamy, A. Ene, and S.A.M. Issa, Sustainability **14**, 3553 (2022).
9. E.S.R. Lasheen, M.K. Azer, A. Ene, W. Abdelwahab, H.M.H. Zakaly, H.A. Awad, and N.A.Kawady, Materials **15**, 4069 (2022).
10. G.A. Alharshan, M.S. Kamar, E.S.R. Lasheen, A. Ene, M.A.M. Uosif, H.A. Awad, S. A. M. Issa, and H.M.H. Zakaly, Int. J. Environ. Res. Public Health **19**, 10717 (2022)
11. H.A. Awad, I. Abu El-Leil, A.V. Nastavkin, A. Tolba, M. Kamel, R.M. El-Wardany, A. Rabie, A. Ene, H.O. Tekin, S.A.M. Issa, and H.M.H. Zakaly, Open Chem. **20**(1), 254–266 (2022).
12. A. Ion, A. Cosac, and V.V. Ene, Appl. Sci. **12**(23), 12363 (2022).
13. A. Yilmaz, H. Yilmaz Alan, Ö. Faydasiçok, L. Amon Susam, R. Şamli, B. Akkuş, A. Erol, E. Güdekli, Ç. İlayda İnci, and M. E. Emirhan, Rom. J. Phys. **67**, 901 (2022).
14. M. Ali, S. Iqbal, M. Wasim, M. Arif, and F. Saif, Radiat. Prot. Dosim. **153**(3), 390–399 (2013).
15. A. Shahrokhi, M. Adelikhah, S. Chalupnik, and T. Kovács, Mar. Pollut. Bull. **163**, 112009 (2021).
16. E. Kapdan, N. Altinsoy, G. Karahan, and A. Yuksel, J. Radioanal. Nucl. Chem. **318**, 1033–1042 (2018).
17. S. Turhan, E. Goren, F. A. Ugur, M. Karatasli, and Z. Yegingil, Radiochim. Acta **106**(2), 161–168 (2018).
18. H. A. Awad, I.A.El-Leil, N. V. Granovskaya, A. Ene, A. Tolba, M. Kamel, A. Nastavkin, R.M. El-Wardany, and H.M. Zakaly, Rom. J. Phys. **67** (5–6), 808 (2022).
19. M.M. El Dabe, A.M. Ismail, M. Metwaly, S.A. Taalab, M.Y. Hanfi, and A. Ene, Materials **15**, 1224 (2022).
20. L.C. Tugulan, O.G. Duliu, F.N. Dragolici, and C. Ricman, Rom. J. Phys. **64**, 805 (2019).
21. S. Dragović, L. Janković-Mandić, R. Dragović, M. Đorđević, M. Đokić, and J. Kovačević, J. Geochem. Explor. **142**, 4–10 (2014).
22. V.G. Linnik, E.M. Korobova, J. Brown, V.V. Surkov, V.N. Potapov, and A.V. Sokolov, J. Geochem. Explor. **142**, 60–68 (2014).
23. A. Ene and A. Pantelică, Rom. J. Phys. **56**(7–8), 1011–1018 (2011).

24. H.A. Saudi, H.T. Abedelkader, S.A.M. Issa, H.M Diab, G.A. Alharshan, M.A.M. Uosif, I.I. Bashter, A. Ene, M.E. Ghazaly, and H.M.H. Zakaly, Int. J. Environ. Res. Public Health **19**, 8124 (2022).
25. R. M. Margineanu, O. G. Duliu, A. M. Blebea-Apostu, C. Gomoiu, and S. Bercea, J. Radioanal. Nucl. Chem. **298**,1191–1196 (2013).
26. A. Ene and A. Pantelică, Radiochim. Acta **98**(1), 53–57 (2010).
27. A. Ene, I. Popescu, V. Ghisa, Rom. Rep. Phys. **61**(1), 165–171 (2009).
28. A. Ene, A. Pantelica, C. Freitas, and A. Boşneaga, Rom. J. Phys. **56** (7–8), 993–1000 (2011).
29. A. Pantelica, C. Freitas, A. Ene, and E. Steinnes, Radiochim. Acta **101**, 45–50 (2013).
30. A. Ene, O. Bogdevich, A. Sion, Sci. Total Environ. **439**, 76–86 (2012).
31. A. Ene, O. Bogdevich, A. Sion, T. Spanos, Microchem. J. **100**, 36–41 (2012).
32. S.S. Moraru, A. Ene, and A. Badila, Sustainability **12**(21), 9174 (2020).
33. C. Tsabaris, G. Eleftheriou, D. L. Patiris, E. G. Androulakaki, N. Kapanadze, F. K. Pappa, G. Melikatze, and G. Ruzsa, J. Radioanal. Nucl. Chem. **321**, 169–181 (2019).
34. M. C. Sandu, G. O. Iancu, C. Chelariu, A. Ion, S. I. Balaban, and A. A. Scarlat, J. Radiat. Res. Appl. Sci. **13**, 730–746 (2020).
35. R.M. Margineanu, A. Ranca, P. Bolos, M.C. Gomoiu, C. Simion, V. Tobosar, and A.M. Blebea-Apostu, Rom. J. Phys. **63**, 809 (2018).
36. C.A. Simion, M. Călin, A.E. Druker, E. Simion, N. Popa, I. Radulescu, N. Mocanu, and S. A. El-Shamali, Rom. J. Phys. **62**, 819 (2017).
37. International Commission on Radiological Protection, *The 2007 recommendations of the International Commission on Radiological Protection*, ICRP Publication 103, Annals of the ICRP **37**(3–4), 53, 2007.
38. M. Tufail, Environ. Monit. Assess. **184**, 5663–5667 (2012).
39. S. M. Darwish, E. Khalifa, M. Al-Abyad, and S. M. El-Bahi, J. Nucl. Radiat. Phys. **11**, 15–35 (2016).
40. S. Dizman, F. K. Gorur, R. Keser, and O. Gorur, Environ. Forensics **20**(3), 211–218 (2019).
41. R. Ravisankar, K. Vanasundari, M. Suganya, Y. Raghu, A. Rajalakshmi, A. Chandrasekaran, S. Sivakumar, J. Chandramohan, P. Vijayagopal, and B. Venkatraman, Appl. Radiat. Isot. **85**, 114–127 (2014).
42. N. M. Antović and N. R. Svrkota, RAP Conference Proceedings **4**, 90–95 (2019).
43. S. Kadiri, S. Dizman, C.M. Yeşilkanat, H. Aliu, and G. Hodolli, Appl. Sci. **12**, 9520 (2022).
44. A. Clouvas, S. Xanthos, M. A. Domis, and J. Silva, Health Phys. **78**(3), 295–302 (2000).
45. K. Mamont-Ciesla, B. Gwiazdowski, M. Biernacka, and A. Zak, *Radioactivity of* building *materials in Poland*, in: K.G. Vohra, U.C. Mishra, K.C. Pillai, and S. Sadavisan (Eds.), *Natural Radiation Environment: Proceedings of the Second Special Symposium on Natural Radiation Environment*, January 19–23, 1981, Bombay, India, John Wiley and Sons Inc., New York, USA, 1982, pp. 551–557.