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Measurement of the radioactivity of ^{238}U, ^{232}Th, ^{226}Ra, ^{137}Cs and ^{40}K in soil using direct Ge(Li) γ-ray spectrometry

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Radioactivity of the nuclides ^{238}U(^{235}U), ^{232}Th, ^{226}Ra, ^{137}Cs and ^{40}K was measured in soil by direct γ-ray spectrometry using Ge(Li) detector. Relative laboratory method was used. Soil was dried, powdered, sieved and put into hermetically sealed container. CCRMP certified reference materials and compounds of the above nuclides mixed with fine quartz sand were used as references. Five and four γ-lines were used for the determination of ^{232}Th and ^{226}Ra, respectively, to obtain more accurate results. The most significant interferences, caused by the limited energy resolution of the detector, were resolved. In the case of ordinary soils, using one day duration of measurement and 1 kg mass of soil, ^{232}Th, ^{226}Ra and ^{40}K can be determined with less than 10% relative random error. Elevated concentrations of ^{238}U(^{235}U) and ^{226}Ra were observed in soil samples collected around a coal-fired power plant in Ajka town, Hungary.

Introduction

Predominant part of the radioactivity of soil derives from the decay of the primordial radionuclides ^{238}U, ^{235}U, ^{232}Th, ^{87}Rb and ^{40}K and the numerous decay products of the first three of them. Significant amounts of man-made radionuclides ^{137}Cs and ^{90}Sr are also present in the soil as a result of nuclear weapons tests and nuclear accidents. The contribution of other nuclides to the total activity of soil is negligible. ^{87}Rb and ^{90}Sr are pure beta emitters so they cannot be determined by γ-ray spectrometry. But their contribution to the total activity is small. The activity concentrations of the nuclides ^{238}U, ^{232}Th and ^{40}K in ordinary soils are about 40, 40 and 400 Bq/kg, respectively.[1]

Direct determination of ^{238}U and ^{232}Th in soil without any chemical treatment using semiconductor γ-ray spectrometer is very hard because ^{238}U and ^{232}Th do not have intensive γ-rays (lines) of their own. But they have several products which have more intensive lines and activities equal to those of their parents in the state of secular equilibrium. In the case of the thorium series, the existence of the equilibrium can be assumed throughout the series for any soil and rock if the emanation is prevented. ^{226}Ra, member of the uranium series, is also in the state of equilibrium with its progeny if the sample is hermetically sealed. But ^{226}Ra is generally not in equilibrium with ^{238}U because of geochemical reasons. ^{238}U has only two decay products which are surely in equilibrium with their parent: ^{234}Th and ^{234m}Pa. ^{235}U/^{238}U isotopic ratio can be assumed to be constant for soils and rocks at $7.2 \cdot 10^{-3}$, so ^{238}U can be determined also through the determination of ^{235}U.

It is usual in the literature in case of ^{232}Th and ^{226}Ra that the nuclide is determined from only one line which is thought to be free from interferences from lines of other nuclides. It is still frequent in the practice that ^{238}U is determined from one line of ^{226}Ra assuming equilibrium. Some authors use the 63.3 keV (3.6%) and 92.6 keV (4.9%) lines of ^{234}Th to determine ^{238}U, but the possibility of using these lines is restricted by the interference of the 92.6 keV line from X-rays of Bi, Po, U and Th,[2] the interference of the 63.3 keV line from the thorium series[3] and the significant self absorption of both γ-rays which depends strongly on the composition of the sample. Ge(Li) detectors, which have lower efficiency and worse energy resolution than modern HPGe detectors do, provide less possibilities and worse performance in the above respect. However, possessing only a Ge(Li) detector, there are still some possibilities to obtain valuable results if some further efforts are made.

A laboratory method has been developed by the authors to determine the activity concentrations of the nuclides ^{238}U, ^{232}Th, ^{226}Ra, ^{137}Cs and ^{40}K in soils by direct (without any chemical or physical separation or concentration) Ge(Li) γ-ray spectrometry. The method is a relative one: certified rock powders and some compounds of the above nuclides mixed homogeneously with analytically clean quartz sand have been used as references. Five and four lines were used for the determination of ^{232}Th and ^{226}Ra, respectively, to obtain more accurate results. The significant interferences from other γ-rays were resolved by a special experimental procedure. The details of the method and some preliminary measurement results obtained for soil samples collected next to a coal-fired power plant in Ajka town, Hungary, are described below.

Experimental

Sample preparations and measurement

The soil was dried, powdered, sieved and poured into hermetically sealed containers. Two types of containers

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were used. The first one was a 110 cm^3 disc-shaped metal tin consisting of a cylindrical lower part and a lid. The lid was fastened to the lower part using an appropriate glue. The other was a 770 cm^3 Marinelli beaker. The lower part of it was made from one piece of Al plate of 1.5 mm thickness. The lid was made from thick Al plate and fastened to the edge of the lower part using 6 screws. The airtight sealing was realized by an "O" ring. 110–150 g and 800–1000 g mass of soil was contained by the tin container and the Marinelli beaker, respectively. The tin container was used in those cases when the mass of the sample was limited.

The γ-ray spectra were measured by a 45 cm^3 coaxial Ge(Li) detector. The energy resolution was 2.7 keV (FWHM) at 661.6 keV. The spectra were evaluated by a Canberra 20 analyzer. The counting rate, $CR = PA/t$, where PA is the net peak area of the full energy peak and t is the duration of the measurement, was measured for the lines of the nuclides to be determined. Dead-time losses were measured by the analyzer. The losses due to the random pile-up were not measured but they were less then about 5% in all cases. The samples were measured more than 30 days after the sealing in order to reach the secular equilibrium between ^{226}Ra and its progeny, ^{214}Pb and ^{214}Bi.

Evaluation of the measurement data

The basic principle was the comparison of the counting rates measured for a given peak of the spectrum (1) in case of a sample containing unknown activity concentration, a, of some radionuclide and (2) in case of the reference sample containing known activity concentration, a_0, of the same nuclide, respectively. The same geometry (fixed volume of sample material) was used, so the full energy peak efficiency was the same in both cases. The relative loss in CR caused by the true coincidences was the same too.

The mass of the sample varied because of the varying density of the sample material. This is why CR was corrected by the factor $K = (m_0/m)\,[\mu d/(1 - \exp(-\mu d))]$, where m is the mass of the sample, m_0 is equal to 100 g and 1000 g for tin and Marinelli beaker, respectively, μ is the mass absorption coefficient of the sample material for the given γ-ray energy in cm^2/g units and d is the average thickness of the sample in g/cm^2 units. The μ values evaluated by HUBBELL[4] for SiO_2 were used, but the contribution of the coherent scattering, σ_{coh}, was estimated from the work of STORM and ISRAEL[5] and subtracted at low energies. The average atomic number of rocks and soils varies within tight bounds and μ depends only slightly on the atomic number above 150 keV. Hence, the same μ was used for all samples. d varied around 3 g/cm^2. It was estimated that the variation of the elemental composition of the sample material caused less than a few percent random error in the final results.

The ratio of the corrected (for background, dead time and different masses) counting rates measured for the unknown and reference samples, CR/CR_0, is then equal to the ratio of the activity concentrations of the given radionuclide in the samples, a/a_0. The unknown difference in the random pile-up rate caused less then a few percent error in the results.

The nuclear data of the γ-rays used for the determination of the above radionuclides can be seen in Table 1. The most intensive lines, except in two cases, were used in order to obtain significant peak areas within an acceptable duration (one day). The most intensive lines among those emitted by ^{234}Th and ^{234m}Pa

Table 1. Energies, intensities and emitter nuclides of γ-rays used for the determination of the different radionuclides

Nuclide to be determined	Peak No.*	Gamma-ray energy, keV	Gamma-ray intensity, %	Emitter nuclide	Reference
^{238}U	1	185.7	57.5	^{235}U	6
^{232}Th	2	238.4	0.11	^{208}Tl	7
		238.6	44.6	^{212}Pb	
		241.0	3.95	^{224}Ra	
	4	338.3	12.4	^{228}Ac	
		341.0	0.40	^{228}Ac	
	6	583.2	30.4	^{208}Tl	
	9	911.2	27.7	^{228}Ac	
	10	964.8	5.2	^{228}Ac	
		969.0	17.3	^{228}Ac	
^{226}Ra	3	295.2	18.5	^{214}Pb	8
	5	351.9	35.8	^{214}Pb	
	7	609.3	44.8	^{214}Bi	
	11	1120.3	14.8	^{214}Bi	
^{137}Cs	8	661.7	85.1	^{137m}Ba	9
^{40}K	12	1460.8	10.7	^{40}K	10

*The peaks are numbered according to the increasing γ-ray energy.

Table 2. The interferences and the nuclear data of the interfering γ-rays

No. of interference	No. of peak*	Interfering γ-ray			
		Energy, keV	Intensity, %	Emitter nuclide	Reference
1	1	186.1	3.50	^{226}Ra	11
2	2	242.0	7.50	^{226}Ra(^{214}Pb)	8
		236.0	11.2	^{235}U(^{227}Th)	10
3	3	300.1	3.28	^{232}Th(^{212}Pb)	12
4	6	580.2	0.35	^{226}Ra(^{214}Pb)	8
5	8	665.5	1.29	^{226}Ra(^{214}Bi)	8
6	10	964.1	0.38	^{226}Ra(^{214}Bi)	8

*Numbered according to Table 1.

are the 63.3 keV and the 92.6 keV ones of ^{234}Th. But because of the problems mentioned in the introduction, these lines were not used and ^{238}U was determined through the determination of ^{235}U using its own intensive line at 185.7 keV (57.5%). The 2614.5 keV (35.8%) line of ^{232}Th(^{208}Tl) was not used because of technical reasons. In case of ^{232}Th and ^{226}Ra, where five and four peaks were used, respectively, the activity was independently determined from all peaks and then the weighed average of the results was computed. This procedure gave more accurate result with lower error as if only one of the peaks would have been used.

A difficulty was caused by the limited energy resolution of the detector. Some of the lines had interferences from other γ-rays and the individual peak areas belonging to the single γ-rays could not be directly determined. These interferences are listed in Table 2. In addition to those listed, there are also some other less important interferences from weak γ-rays of the thorium and actinium series, but these may cause at most only 1-2% error in the final results. In all cases listed in Table 2, only the area of the complex peak was measured. The interferences were resolved using the ratios between different peaks. To separate the contribution of ^{235}U in the complex 185.7 + + 186.0 keV peak, for example, firstly a sealed sample containing only ^{226}Ra was prepared and the *PA* (net peak area) ratios of the peaks 186.0/295.2, 186.0/351.9, 186.0/609.3 and 186.0/1120.3 were determined. Then *PA* of the 295.2, 351.9, 609.3 and 1120.3 keV peaks for the unknown sample were measured and multiplied by turns with the corresponding one of the above ratios. These products all gave the probable value of the $PA_{186.0}$ contribution of ^{226}Ra in the area of the complex peak for that sample because the above *PA* ratios depend only slightly on the elemental composition of the sample. $PA_{186.0}$ was computed as the weighed average of the four products and subtracted from the total peak area to obtain $PA_{185.7}$.

Similar procedures were used also in the other cases listed in Table 2. The sequence of these procedures was fixed. For example, $PA_{295.2}$ was computed before computing $PA_{185.7}$. The following sequence was applied (according to the number of the interferences): 4→6→3→1→2→5. Of course, only separated peak areas were used in every step. For example, in steps 4 and 6 $PA_{295.2}$ was not used.

Table 3. Activity concentrations of ^{238}U, ^{226}Ra and ^{232}Th in the reference materials

Name	Source of data	Activity concentration ± 95% confidence level,* Bq/kg		
		^{238}U	^{226}Ra	^{232}Th
DH-1a	CCRMP	32 450 ± 40	31 500 ± 1100	3680 ± 120
	This work	31 810 ± 1000	31 350 ± 140	3600 ± 50
DL-1a	CCRMP	1430 ± 40	1400 ± 40	310 ± 15
	This work	1440 ± 110	1400 ± 10	310 ± 2
BL-4a	CCRMP	15 400 ± 90	15 500 ± 500	–
	This work	15 600 ± 600	15 450 ± 60	70 ± 8
SY-3	CCRMP	8020	7470 ± 370**	4000
	This work	8540 ± 400	8920 ± 40	4080 ± 40
DT-4	MOE	6800	6680	8900
	This work	6200 ± 350	6000 ± 70	7100 ± 100
DU-1	MOE	385 900	382 000	40
	This work	455 600 ± 22 800	417 000 ± 2250	< 100

*Confidence levels were not given by the producer for SY-3, DT-4 and DU-1.

**Published by GLADNEY et al.[15]

Reference samples

One main source of systematic error in using relative method is that the certification of the material used as reference may have some uncertainty. In this work a few high quality reference materials were used for the determination of ^{238}U(^{235}U), ^{232}Th and ^{226}Ra to obtain reliable results. Four reference samples certified by the Canadian Certified Reference Materials Project (CCRMP) (namely DH-1a, DL-1a, BL-4a and SY-3) were used. Two other materials produced by the Mecsek Ore Enterprise (MOE), Hungary (namely DT-4 and DU-1) were used as auxiliary references. The a_0 activity concentration data of the original materials provided by the producers can be seen in Table 3. The materials having very high activity concentrations of ^{238}U, ^{226}Ra and ^{232}Th (DH-1a, DT-4 and DU-1) were diluted with analytically clean fine quartz sand. Some further reference materials were made by the authors mixing homogeneously known amounts of U_3O_8, $Th(NO_3)_4$, $(Ra,Ba)SO_4$, KCl and $^{137}CsNO_3$ into analytically clean fine quartz sand. Uranium in U_3O_8 had natural isotopic composition, $Th(NO_3)_4$ was produced in 1906 and supplied by Amersham International. The activity concentrations of ^{226}Ra in $(Ra,Ba)SO_4$ and of ^{137}Cs in $^{137}CsNO_3$ have been determined by the authors through the measurement of point sources made from these compounds. Previously the detector was calibrated for point-source geometry. The emanation of ^{222}Rn from the crystals of $(Ra,Ba)SO_4$ was negligible, so the equilibrium between ^{226}Ra and its progeny to ^{214}Po was certain in this compound.

After measuring CR_0 for the reference samples in tin geometry, the counting rate divided by the activity concentration of the emitter nuclide, $s_0 = CR_0/a_0$, was computed for all the reference samples, nuclides and lines.

As to ^{226}Ra, the s_0 values obtained were almost the same for DH-1a, DL-1a, BL-4a and mixed $(Ra,BA)SO_4$ for all lines within a standard deviation of 3%. The deviations of SY-3, DT-4 and DU-1 from the average of the above four were +19%, −10% and +9%, respectively. a_0 of ^{226}Ra in DH-1a, DL-1a and BL-4a was certified on the basis of the measurements of the same 13 laboratories,[13–14] so the good agreement is easily understood. For SY-3 the a_0 value published by GLADNEY et al.[15] was used because CCRMP did not report result on ^{226}Ra. The large deviation obtained for SY-3 can be explained perhaps with the inhomogeneity of the material, the possibility of which was not precluded by CCRMP.[16] Because of the very good agreement among mixed $(Ra,Ba)SO_4$, DH-1a, DL-1a and BL-4a, the authors decided to accept the averages of the s_0 values obtained for the four references as the best approaches for the lines of ^{226}Ra.

Table 4. Counting rate from a unit activity concentration, s_0, for $m_0 = 100$ g and 1000 g mass of sample for tin and Marinelli-beaker geometries, respectively

Nuclide to be determined	No. of peak (energy, keV)	$s_0 \pm \Delta s_0$,* (ks^{-1})/(Bq · kg^{-1})	
		Tin	Marinelli beaker
^{238}U	1(186)	0.0595 ± 0.0005	0.353 ± 0.011
^{232}Th	2(239)	0.815 ± 0.019	4.946 ± 0.027
	4(338)	0.109 ± 0.003	0.716 ± 0.004
	6(583)	0.156 ± 0.001	0.952 ± 0.004
	9(911)	0.0811 ± 0.0003	0.531 ± 0.010
	10(967)	0.0551 ± 0.0025	0.386 ± 0.004
^{226}Ra	3(295)	0.241 ± 0.002	1.477 ± 0.010
	5(352)	0.381 ± 0.002	2.337 ± 0.010
	7(609)	0.225 ± 0.002	1.400 ± 0.007
	11(1120)	0.0362 ± 0.0001	0.223 ± 0.006
^{137}Cs	8(662)	0.436 ± 0.009	2.625 ± 0.014
^{40}K	12(1461)	0.0196 ± 0.0001	0.120 ± 0.0002

*1 σ error of the mean.

In case of the 185.7 keV line of ^{238}U(^{235}U), the s_0 values obtained for DH-1a, DL-1a, BL-4a and mixed U_3O_8 agreed well within 4%, while SY-3, DT-4 and DU-1 deviated from the above four by +6%, −9% and +18%, respectively. a_0 of ^{238}U in DH-1a, DL-1a and BL-4a was certified on the basis of the measurements of 9, 20 and 1 laboratories, respectively.[17–19] Because of the good agreement among mixed U_3O_8, DH-1a, DL-1a and BL-4a, the average of the s_0 values obtained for the above four reference samples was accepted as the best approach for the 185.7 keV line.

For ^{232}Th, the s_0 values obtained were the same within 5% for DH-1a, DL-1a, SY-3 and mixed $Th(NO_3)_4$ for all lines. The deviation of DT-4 from the rest was −20%. The ^{232}Th content of DH-1a, DL-1a and SY-3 were certified in partly independent interlaboratory programs with 13, 18 and 17 participating laboratories.[16–18] Because of the good agreement between mixed $Th(NO_3)_4$, DH-1a, DL-1a and SY-3, the authors decided to accept the averages of the s_0 values obtained for the four reference samples as the best approaches for the lines of ^{232}Th.

The best approaches for the s_0 values evaluated by using the above procedure can be seen in Table 4. After their evaluation, activity concentrations for a sample having unknown radionuclide composition were computed as $a = CR/s_0$. This was applied also for the reference materials. The values so obtained can also be seen in Table 3.

Similar procedures were applied for Marinelli beakers using DT-4, DU-1 (diluted with fine quartz sand), $(Ra,Ba)SO_4$, KCl and $^{137}CsNO_3$ as standard samples and a_0 values obtained above. The s_0 values obtained for Marinelli-beaker geometry can also be seen in Table 4.

Results and discussion

Performance of the method

The existence of the equilibrium between ^{226}Ra and its progeny in some rock-type reference materials in tin container were controlled by determining the ratio $PA_{185.7}/PA_{186.0}$, where $PA_{186.0}$ was estimated according to the above from the lines of ^{214}Pb and ^{214}Bi. The values obtained for DH-1a, DL-1a and BL-4a are 0.77 ± 0.01,

Table 5. Relative statistical error (1σ) of the activity concentration, Δα, for a soil having average radioactivity (40, 40, 40 and 400 Bq/kg for the nuclides ^{238}U, ^{232}Th, ^{226}Ra and 40K, respectively) and for one day duration of measurement and the lower detection limits (LDL) for the activity concentration (above which the statistical error decreases below 50%)

Nuclide	Δα (ordinary soil, 1 day duration of measurement), %		LDL for activity concentration, Bq/kg	
	Tin	Marinelli beaker	Tin	Marinelli beaker
^{238}U	90	35	75	25
^{232}Th	5	2	4	2
^{226}Ra	6	2	3	2
^{137}Cs	–	–	4	2
^{40}K	9	3	50	20

Table 6. Activities of some soil samples collected next to the Ajka coal-fired power plant

Origin*	Depth, cm	Geometry**	Activity concentration, Bq/kg				
			^{238}U	^{226}Ra	^{232}Th	^{137}Cs	^{40}K
Brook-side	0–5	T	343 ± 66	404 ± 6	31 ± 5	31 ± 2	284 ± 29
	5–10	T	443 ± 64	532 ± 7	34 ± 4	9 ± 3	256 ± 42
	10–15	T	233 ± 52	183 ± 7	34 ± 3	7 ± 3	337 ± 39
	15–20	T	< 100	86 ± 3	35 ± 3	4 ± 2	305 ± 35
	20–25	T	< 100	69 ± 3	42 ± 3	4 ± 2	325 ± 38
Street-corner	0–5	T	199 ± 56	412 ± 7	19 ± 4	58 ± 4	235 ± 45
	5–10	T	271 ± 59	421 ± 6	21 ± 4	18 ± 4	258 ± 45
	10–15	T	418 ± 91	469 ± 8	30 ± 6	17 ± 4	179 ± 57
	15–20	T	944 ± 170	798 ± 14	< 20	25 ± 7	239 ± 88
	20–25	T	420 ± 69	607 ± 9	21 ± 5	11 ± 4	252 ± 49
Street-side	0–5	T	86 ± 36	62 ± 4	23 ± 3	11 ± 2	332 ± 34
	5–10	T	< 90	54 ± 3	23 ± 2	8 ± 2	327 ± 32
	10–15	T	< 40	20 ± 2	22 ± 2	4 ± 1	292 ± 28
Hen-yard	0–20	M	492 ± 79	485 ± 7	20 ± 4	69 ± 4	351 ± 41
Kitchen-garden	0–20	M	122 ± 20	97 ± 2	24 ± 1	19 ± 1	330 ± 15
Pit-bottom	100–110	M	33 ± 13	30 ± 1	30 ± 1	< 2	448 ± 30
Plough-land	0–20	M	34 ± 11	50 ± 1	39 ± 1	14 ± 1	413 ± 11
Fallow-land	0–20	M	41 ± 16	36 ± 1	31 ± 1	11 ± 1	333 ± 11

*First 6 are from inside the town, last 2 are from outside the town within a few kilometers.

**T = tin container; M = Marinelli beaker.

0.78 ± 0.03 and 0.75 ± 0.01, respectively. The efficiency and the absorption are the same for the 185.7 keV and 186.0 keV γ-rays. Hence, these ratios can be computed also from the nuclear data published in the literature.[6,11,20] the ^{235}U/^{238}U isotopic ratio and the a_0 data provided by CCRMP. In this way 0.774, 0.767 and 0.747 values can be obtained. The excellent agreement between the measured and computed values shows that the sealing of the tin container is perfect.

The relative statistical errors of the activity concentration results for an ordinary soil sample and for one day duration of measurement can be seen in Table 5. Some further random error of at most 5% comes from the variation of the unknown random pile-up and elemental composition. The lowest activity concentrations which can be measured by this method (above which the statistical error decreases below 50%) can also be seen in Table 5. A further systematic-type error of about 5% can be assumed from the uncertainties of the calibration procedure and the unresolved little interferences.

The random error for ^{238}U in case of an ordinary soil is rather high. More accurate results can be obtained for it in case of soils having elevated uranium content. All the other radionuclides can be determined with sufficient accuracy for many practical purposes. The results depend only slightly on the elemental composition of the sample through the value of the mass absorption coefficient, μ. Hence, the method can be applied without correction for materials having similar elemental composition, such as most rocks and some building materials. The method can be applied even for materials having different composition using appropriate correction taking the different value of μ into account.

In addition, the method provides a possibility for the determination of the emanation coefficient of the sample material. During the process of sample preparation, ^{222}Rn may leave the sample without difficulty and a of ^{214}Pb and ^{214}Bi decreases to that level which is in equilibrium with the proportion of ^{222}Rn remaining enclosed in the grains of the sample material. Measuring a spectrum next after the sample preparation, a obtained from the lines of ^{214}Pb and ^{214}Bi will not be equal to a of ^{226}Ra but will be equal to that of the enclosed ^{222}Rn. Dividing this value of a with that measured for ^{226}Ra after a 30 days delay and subtracting the result from the unity, the emanation coefficient can be obtained.

Preliminary results obtained for soil samples collected in Ajka, Hungary

Soil samples were collected in Ajka town, Hungary. Ajka is a mining and industrial town lying in the Bakony Mountain, 30 km north of Lake Balaton. Cretaceous brown coal rich in uranium is mined by the town since 1865. The average activity concentration of uranium in this coal nowadays is about 300–500 $\mathrm{Bq \cdot kg^{-1}}$ but reached the value of 800–900 $\mathrm{Bq \cdot kg^{-1}}$ 40 years ago. At the same time, the thorium content of the coal is not high. (The geohistorical explanation was given by SZALAY).[21]

A huge amount of the above coal has been used up in the town for heating and industrial energy production since 1865. The biggest consumer of the coal is now a coal-fired power plant established in 1943 and settled inside the border of the town. The electrical energy generated by the plant increased in a few steps from 20 to 90 MW · a. The radioactivity of the coal was not known before the fifties. In the sixties the authorities prohibited the use of slag of Ajka coal by the building industry. However, before that time and slightly even after that the slag was used in the town for filling the irregularities of the ground. The chimneys of the plant were not equipped with effective filters before the end of the eighties.

The samples were collected in the town and in the neighbourhood of the town from about 80 different places. The samples were investigated by the above method. The results of this work and the conclusions will be published in detail in another forthcoming paper. A few interesting results are listed in Table 6. The emanation coefficient for 17 samples was varied between 0.12 and 0.40 with an average and standad deviation 0.28 ± 0.10. Our main experience was that the concentration of ^{238}U and ^{226}Ra increased significantly in the upper layers of the soil compared with the original values. The main reason of this over and above the deposition of the fly ash is the mixing of the original soil with the slag of the coal due to industrial building activity and some household activities of the private population.

Conclusions

It has been shown that even using a Ge(Li) detector of average performance, valuable measurement results can be obtained if the information beeing present in the γ-spectra is better exploited. The use of a few high quality reference materials made the method more accurate and reliable The method can be used to solve many practical radioanalytical problems related to the materials having similar composition to that of the earth crust.

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