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A DETERMINATION OF THE CONCENTRATION LEVEL OF LEAD ²¹⁰Pb ISOTOPE IN SOLID SAMPLES FOR THE ASSESSMENT OF RADIATION RISK OCCURRING IN COAL MINES

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Abstract

Lead ²¹⁰Pb, an element of the natural uranium radioactive decay series, is not currently considered a source of radiation risk, especially in a radiation protection system in underground mines in Poland. However, it could be a completely independent element of the radioactive series due to its physical and chemical properties. Routine measurements showed significantly higher than expected concentrations of ²¹⁰Pb in underground radium rich sediments, based only on the radioactive decay law. This phenomenon implies a need of ²¹⁰Pb concentration monitoring in such sediments. Nevertheless, the laboratory analysis of ²¹⁰Pb by gamma radiation spectroscopy is connected with a particular hindrance, the self-attenuation of ²¹⁰Pb radiation in samples. Current work describes a practical method for obtaining the self-attenuation correction factor in the case of ²¹⁰Pb concentration analysis. Experimentally obtained correction factors range between 0.51–6.96 cm²/g. Neglecting this factor can cause a significant error or underestimations in radiological risk assessment.

Keywords

self-absorption, correction factor, gamma spectrometry, lead Pb-210

1. INTRODUCTION

Lead ²¹⁰Pb is a naturally occurring radioactive nuclide element of the uranium (²³⁸U) radioactive series. It is produced as a result of the decay of so-called short-lived progeny of ²²²Rn, which comes from the decay of ²¹⁴Po (99.98%) and ²¹⁴Bi by ²¹⁹Tl (0.02%). ²¹⁰Pb is subject to beta decay into ²¹⁰Bi. This process is accompanied by the emission of a quantum of gamma radiation of energy 46.5 keV. ²¹⁰Bi is subject to beta decay which results in ²¹⁰Po being produced, which is subject to alpha decay into a stable isotope, ²⁰⁶Pb. This transformation is accompanied by the emission of alpha particle type. Therefore, the analysis of concentration ²¹⁰Po requires complicated measuring methods based on its chemical separation (Clayton, Bradley 1995). On the other hand, ²¹⁰Po, as an alpha-radioactive nuclide, is extremely dangerous when absorbed by the human body. The values of dose coefficient for a few selected nuclides of uranium series is presented in Table 1.

Table 1. Dose coefficient e.g. for employees who ingested or inhaled some nuclides into their bodies

Nuclide	Inhalation		Ingestion e(g) Sv/Bq
	e(g) 1µm Sv/Bq	e(g) 5µm Sv/Bq	
²¹⁰ Pb	8.9·10 ⁻⁷	1.1·10 ⁻⁶	6.9·10 ⁻⁷
²¹⁰ Bi	1.1·10 ⁻⁹	1.4·10 ⁻⁹	1.9·10 ⁻⁹
²¹⁰ Po	6.0·10 ⁻⁷	7.1·10 ⁻⁷	2.4·10 ⁻⁷

The mutual relationship of the half-life time ²¹⁰Pb, ²¹⁰Bi and ²¹⁰Po result in these nuclides reaching a state of secular radioactive equilibrium relatively quickly (Fig. 1). This fact should be taken into account in each case of radiation risk assessment or the assessment of environmental impact. Moreover, it can be used (or constitute a limitation) in the measurement methods of ²¹⁰Pb.

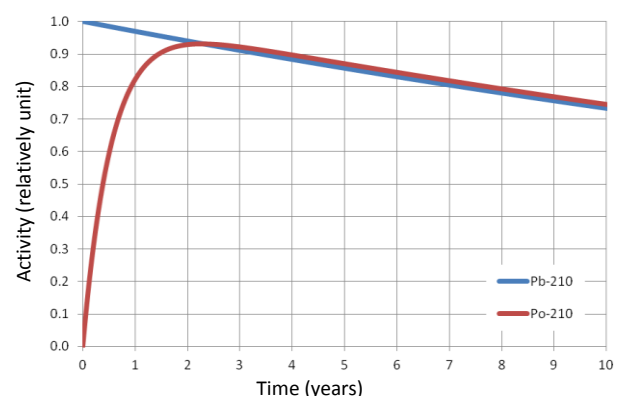


Figure 1. Changes of concentration in time ²¹⁰Pb and ²¹⁰Po in an isolated system (state of secular radioactive equilibrium between ²¹⁰Pb and ²¹⁰Bi is reached after about 50 days)

Due to a relatively long half-life time (22.3 years) ²¹⁰Pb is the sole natural radioactive lead isotope, the presence of

which in the environment is directly related to the presence of the parent isotope. Its concentration can vary depending on natural environmental processes (atmospheric, hydrological) or anthropogenic.

The most important factors of radiation risk in mines include radioactive mine water and sediment formed from it which contains increased concentrations of radium isotopes (^{226}Ra and ^{228}Ra). They are formed by the co-precipitation of radium with barium sulphate and the subsequent sedimentation. Since observations of ^{210}Pb in mine water concentration is generally very low, it is assumed that it is not involved in the process of sediment formation, and therefore it should not be found in these sediments at the time of their formation. Its accumulation is related to radium decay, in accordance with the law of radioactive decay (Michalik 2011).

Based on the measurements of the concentration level of radioactive nuclides in the sediments, carried out under the routine control of radiation hazard in mines, it can be concluded that the observed concentration rarely corresponds to the value resulting only from the decay of radium. Among the sediments examined in the Laboratory of Radiometry in the Central Mining Institute – GIG (now: Silesian Centre for Environmental Radioactivity in the Central Mining Institute – GIG) over the past 20 years it has been noted that in approximately 70% of cases, the radioactive concentration of ^{210}Pb significantly exceeded the value resulting from the decay of only ^{226}Ra . In about 22% of examined sediments the correspondence of lead concentration with this value was found, while in nearly 8% of cases ^{210}Pb concentration turned out to be much lower than the expected value. This means that for the majority of formed sediments the accumulation of ^{210}Pb takes place not only as a result of the decay of ^{226}Ra , but also as a result of previously unrecognized geochemical processes.

The observed concentrations of ^{210}Pb in sediments precipitating from mine water can even reach several dozen becquerels per kilogram of sediment mass. By comparison, the typical values of the concentration of ^{210}Pb in soil in Poland averages from 7.9 to 91.2 becquerels (Bq) per kilogram (Radiologiczny... 1997). Such a high amount of lead is a significant contribution to the effective dose received by staff working in mines in the immediate vicinity of these sediments (mainly as a result of accidental inhalation or ingestion). On the other hand, the high number of samples, in which the concentration of lead is considerably lower than the expected values based on the decay of radium, indicates the extreme mobility of this nuclide.

In connection with the above there is a danger of it escaping into the environment from areas of sediment deposition (landfills) or directly from settlers located on the surface, which most frequently have not been isolated effectively enough from the environment. The risk of this isotope migrating becomes quite significant as it could find its way into mine water which could result in the contamination of drinking water. Because of its chemical properties, lead cannot be removed from the human body. After entering the human body a relatively rapid rise of ^{210}Po ought to be expected, which is characterised by a high value of dose coefficient. As a result, penetration of each becquerel of ^{210}Pb isotope by ingestion (or inhalation), for instance as a result of consumption, can lead to a significant increase in exposure to radiation in terms of radiation protection requirements. Therefore, it is

necessary to determine ^{210}Pb accurately in different environmental samples.

The physical and chemical properties of lead and the parameters of decay of ^{210}Pb isotope is a cause of difficulty for the accurate determination of this nuclide, in particular in samples, which vary considerably in density and chemical composition. This is significant in the case of sediment which is produced from mine water. Below, there are three analytical methods for the determination of ^{210}Pb concentration in samples.

2. A REVIEW OF THE RESEARCH METHODS

During the decay of the atom of lead 210, beta and gamma radiation is emitted. As mentioned above, the state of secular radioactive equilibrium of lead and polonium, which is the emitter of alpha radiation, is relatively quick to define. Thus, for the measurement of ^{210}Pb concentration, measurement methods can be used which are based on the detection of each type of ionizing radiation.

It is possible to use gamma ray spectrometry in tested materials, where there is a relatively high activity of ^{210}Pb . A typical detection limit for this method is about 0.4 Bq/kg. The application of shields made of lead which do not contain ^{210}Pb isotope, allows the background to be lowered and thus improve the detection limit. However, such solutions are extremely expensive. Additional difficulties when measuring by means of gamma radiation spectrometry are connected with the low energy of radiation emitted by ^{210}Pb . This necessitates the use of appropriate corrections related not only to the density but also to the chemical composition of the matrix.

One of the methods to estimate the size of the correction is to use the Monte-Carlo simulation, based on the knowledge of the chemical composition (Jodłowski 2006). The main disadvantage of this solution is the need to know the chemical composition, this increases the cost and time needed for the study. An alternative to this solution (which also requires prior knowledge of the chemical composition) is the application of a standard sample of chemical composition and density similar to the examined sample. A different solution, widely used in laboratories, is to estimate the size of self-absorption with the use of the transmission experiment (Cutshall, Larsen, Olsen 1983). After the measurement and initial determination of ^{210}Pb activity, a point source of ^{210}Pb of known activity is placed over the tested sample located in the radiation detector. In order to reduce dispersion and to obtain a coherent beam of radiation perpendicular to the surface of the sample, a collimator is used. Then, the intensity of adsorption is determined in the peak corresponding to 46.5 keV and with the use of appropriate formulas, the correction is calculated. The main advantage of this solution is a lack of necessity to determine the chemical composition of a tested sample. However, one of the disadvantages of this method ought to be mentioned, this is the limitations in the applicable measurement geometry and very simplified mathematical descriptions of the phenomenon of the absorption in the sample, on the basis of which a formula is developed for the calculation of the final correction. The advantage of using gamma radiation spectrometry in general is the lack of necessity for the chemical preparation of samples subject to a measurement and a relatively short time required to obtain test results. Usually it does not exceed 24 hours.

The spectrometry of liquid scintillation counting (LSC) enables the detection of beta radiation, emitted by radioactive isotopes contained in the analysed material, with a considerable, almost close to 100%, efficiency. This method also allows radiation energy to be distinguished, which in turn ensures the identification of isotopes, which are a source of registered radiation. The required condition necessary to carry out the measurement with the use of liquid scintillation counting is to transform the analysed material to a solution.

This solution, when mixed with liquid scintillation, is a sample directly measured in a scintillation counter. A typical limit of detection for this method shapes at the level of several dozen milibecquerels per sample. The use of chemical preparation ensures the extraction of lead and its concentration which provides an even lower detection limit. It comes down to the need for developing a specific procedure for chemical preparation for each matrix. Due to the fact that the currently available liquid scintillation spectrometers are not able to completely separate the signal from the registration of beta radiation of ^{210}Pb and ^{210}Bi , the optimum level of the signal in relation to the background can be achieved after the determination of secular radiation equilibrium between isotope ^{210}Pb and ^{210}Bi .

The result is that the waiting time for measurement results is extended to nearly a month from the termination of chemical preparation. The measurement can be done before, but in such a situation an identical time regime ought to be used for examined samples and standard samples used for calibration, this greatly complicates the measurement cycle, and as a result does not provide the optimum limit of detection. Low energetic resolution of liquid scintillation spectrometers also necessitates the use of chemical preparation, this guarantees the removal of the sample from other radioactive nuclides, which are the emitters of beta radiation, the presence of which in the analysed sample is not in any way associated with the presence of ^{210}Pb isotope. This applies mainly to the isotopes of radium and thorium, and also their derivatives, particularly those that occur in the thorium series.

The measurement of the concentration of ^{210}Pb can also be performed by means of a spectrometry of alpha radiation, using the stabilization of radioactive equilibrium between ^{210}Pb and ^{210}Po (Vajda et al. 1997). This method, supplemented by relevant chemical preparation, enables the concentration of lead at a lower level than the method of liquid scintillators to be determined. Its drawback is the need to know the current ratio of the isotope ^{210}Pb to the isotope ^{210}Po in the examined sample, as the existence of the state of permanent radiation equilibrium between these nuclides cannot always be assumed, particularly in materials of biological origin. The safest solution is to carry out a polonium extraction from the examined sample after 5 half-value periods of ^{210}Po isotopes from its collection, which amounts to almost two years. Such a long time to wait for a test result is not acceptable for many reasons.

3. THE METHOD USED IN THE SILESIAN CENTRE FOR ENVIRONMENTAL RADIOACTIVITY IN THE CENTRAL MINING INSTITUTE

In the Silesian Centre of Environmental Radioactivity, ^{210}Pb is routinely determined in solids by the method of gamma ray spectrometry. As indicated previously, the main

limitation of this method is the phenomenon of self-absorption of low-energy gamma radiation in the material of the sample. This phenomenon is mainly due to photoelectric absorption and Compton scattering. The intensity of self-absorption increases in proportion to the density of the sample and the content of heavy elements (thus with the effective atomic number). Consequently, gamma radiation, emitted by nuclides of lead contained in the sample, is weakened to the extent depending on the type of material from which the sample was collected. This fact prevents the accurate determination of radioactive concentration of the nuclide based on the performance curve of the detector, prepared on the basis of the different chemical composition than the examined sample. Accordingly, it becomes necessary to determine the corrections for self-absorption for each examined sample.

In the Silesian Centre for Environmental Radioactivity it is determined using the so-called transmission method, based on the measurement of the total mass absorption coefficient. Then, the correction is calculated, which is used to revise the result of ^{210}Pb activity in the sample under analysis, which was obtained as a result of a routine measurement. The available publications provide a few variants of relevant calculation formula (Cutshall, Larsen, Olsen 1980; Khater, Ebaid 2008; Jodłowski 2006). Because of the doubts raised by the correctness of the calculations carried out in the cited works, the exact derivation of the formula for correction C is presented below.

For this study a sample of the material containing ^{210}Pb in the shape of a flat cylinder, which lies directly on a flat detector (Fig. 2) was used. It was assumed that the nuclides of lead are distributed uniformly in the whole volume of the sample.

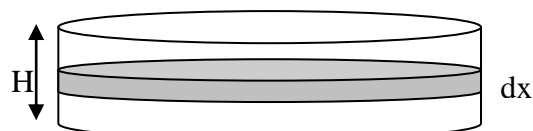


Figure 2. The sample in the cylindrical geometry (scale pan) with a height H

The whole volume of the sample was divided with a plane parallel to the base for i indefinitely small layers of height dx . For each layer separated in this way it can be assumed that due to lead nuclides decay, the number of gamma radiation quanta generated in it (some of which will be self-absorbed) will be equal to:

$$\frac{I_R}{H} dx \quad (1)$$

where:

I_R – the actual number of gamma radiation quanta, generated in the whole volume of the sample (this number is a direct result of the sample's activity)

H – height of the sample

dx – thickness of i -th layer

Classical, the weakening of the beam of gamma radiation quanta in the matter can be described by means of (modified) Lambert-Beer's law:

$$I = I_0 e^{-\mu dx} \quad (2)$$

where:

I_0 – the intensity of the beam of gamma radiation quanta incident on a given medium

I – the intensity of the beam after passing through the medium of density d [$\text{g}\cdot\text{cm}^{-3}$] and thickness x [cm]

μ – mass weakening coefficient [$\text{cm}^2\cdot\text{g}^{-1}$]

According to formula (2) the number of quanta of gamma radiation ($I_{p,i}$) registered in the detector from the i -th layer of the sample located in the distance x from the detector equals:

$$I_{p,i} = \frac{I_R}{H} dx e^{-\mu dx} \quad (3)$$

Totalizing calculated in this way, numbers of quanta of gamma radiation for all i layers, the following is obtained:

$$I_p = \int_0^H \frac{I_R}{H} e^{-\mu dx} dx \quad (4)$$

Solution of the integral (4) is

$$I_p = I_R \frac{1 - e^{-\mu dH}}{\mu dH} \quad (5)$$

Hence

$$C = \frac{I_R}{I_p} = \frac{\mu dH}{1 - e^{-\mu dH}} \quad (6)$$

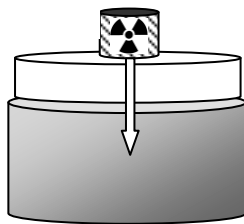


Figure 3. Measuring system comprising a source of ^{210}Pb in the collimator (1), the tested sample in a cylindrical measuring vessel (2) and a germanium detector (3)

Determined on the basis of the above formula, value C is called the correction for self-absorption.

In the transmission experiment (carried out according to Cutshall's method) directly on the sample, a source of gamma radiation is observed with energy, for which corrections ought to be determined, and a number of impulses is calculated and registered in the detector (I_{ZP}). Then, the experiment is repeated after removing the sample so that the source remains at the same distance from the detector, registering the number of impulses (I_{Z0}). Using Lambert-Beer's Law for the second time (2), the following is obtained:

$$I_{ZP} = I_{Z0} e^{-\mu dH} \quad (7)$$

or

$$\frac{I_{ZP}}{I_{Z0}} = e^{-\mu dH} \quad (8)$$

Finding the logarithm to the equation (7) and (8), the following is received:

$$\mu dH = \ln \left(\frac{I_{Z0}}{I_{ZP}} \right) \quad (9)$$

Substituting (8) and (9) into (6), the following is obtained:

$$C = \frac{\mu dH}{1 - e^{-\mu dH}} = \frac{\ln \left(\frac{I_{Z0}}{I_{ZP}} \right)}{1 - \frac{I_{ZP}}{I_{Z0}}} \quad (10)$$

or directly for measurements in reference to the measurement standard:

$$C = \frac{\ln \left(\frac{I_W}{I_P} \right)}{1 - \frac{I_P}{I_W}} \quad (11)$$

where I_W, I_P are the numbers registered in the detector while gammascopying both the standard and the sample.

The determination result received directly from the routine measurement ought to be multiplied by the calculated value of correction C . In the realization of this method, in order to determine the correction for ^{210}Pb , the key element is the use of a highly collimated beam of radiation of energy 46.5 keV. The point source of ^{210}Pb was used with the activity of 208 ($\pm 4\%$) kBq in the Silesian Centre of Environmental Radioactivity. The source has a shape of a flat disc (with dimensions: 25 mm – diameter, 3 mm – height) made of Plexiglas, with a centrally located radioactive material (ca. 1×1 mm). Locked in a lead collimator, it was positioned directly above the sample (Fig. 3).

The tested sample was located in a cylindrical container with dimensions: 10 mm – height, 90 mm – diameter, routinely used for the measurements by means of a gamma ray spectrometry. Adopted for the quantitative analysis, the efficiency curve of the detector was determined on the basis of the measurement standard containing the certified reference material RGU-1, produced by the International Atomic Energy Agency (IAEA) in Vienna, containing the nuclides of uranium radioactive series in secular radioactive equilibrium. The density of the standard material is 1.15 g/cm^3 .

In the experiments, the germanium detector type BEGe (*broad energy*) manufactured by Canberra was used. The detector has a shape of a flat cylinder with a diameter of 81 mm and height 31 mm. The active area of the detector was ca. 5000 mm^2 . Peak resolution (FWHM) for cobalt ^{60}Co source was 2.2 keV – for 1332.5 keV. The resolution for 46.5 keV (^{210}Pb) was typically 0.60 keV.

4. TEST RESULTS

A measurement of the linear absorption coefficient was made and on its basis, correction C was calculated for 60 various samples of mine sediments. The examined samples were characterised by a wide range of radioactive concentration of radium and lead isotopes:

- ^{226}Ra : 41.7–62 230 Bq/kg,
- ^{228}Ra : 16.7–22 250 Bq/kg,
- ^{210}Pb : 56.2–6 040 (without correction) or 42.2–11 700 (with correction) Bq/kg.

The density of the examined samples ranged within: $0.45\text{--}1.40 \text{ g/cm}^3$. The detailed results of the obtained measurements are shown in Table 2.

Table 2. The results of the measurements of the correcting factor

No.	²²⁶ Ra Bq/kg	²²⁸ Ra Bq/kg	²¹⁰ Pb Bq/kg	²¹⁰ Pb* Bq/kg	Density g/cm ³	C	C** cm ² /g
1	17 190 ±440	72.9 ±9.9	6040 ±320	8330 ±610	0.99	1.38	1.10
2	2755 ±72	44 ±12	2110 ±120	1920 ±150	0.96	0.91	0.74
3	1531 ±39	56.9 ±4.4	1249 ±58	1199 ±82	0.98	0.96	0.77
4	850 ±26	44.3 ±8.8	896 ±69	725 ±67	0.86	0.81	0.74
5	616 ±19	36.7 ±8.7	607 ±59	491 ±54	0.89	0.81	0.71
6	443 ±12	38.9 ±2.3	355 ±17	273 ±19	0.96	0.77	0.63
7	950 ±28	48.2 ±4.3	758 ±42	598 ±45	0.82	0.79	0.76
8	2018 ±19	40.9 ±5.1	1621 ±77	1590 ±110	1.08	0.98	0.72
9	348 ±9	31.9 ±2.2	327 ±19	297 ±23	1.21	0.91	0.59
10	230 ±8	34.1 ±2.7	249 ±16	209 ±18	0.86	0.84	0.77
11	2692 ±69	39.2 ±6.3	1520 ±80	1380 ±110	0.97	0.91	0.74
12	1497 ±37	32 ±3.4	1015 ±47	954 ±66	1.06	0.94	0.70
13	747 ±20	26.4 ±4.1	502 ±34	416 ±36	1.07	0.83	0.61
14	5530 ±150	47.9 ±5.6	3230 ±150	3130 ±210	0.94	0.97	0.81
15	12 800 ±300	89 ±22	4640 ±220	5710 ±390	1.13	1.23	0.85
16	301 ±8	19.5 ±1.7	214 ±13	192 ±16	1.38	0.90	0.51
17	664 ±21	33.9 ±5.9	719 ±46	560 ±46	0.75	0.78	0.82
18	557 ±16	40.2 ±3.2	642 ±22	526 ±32	0.84	0.82	0.77
19	91.5 ±3.1	29.4 ±2.9	133 ±11	102 ±10	0.71	0.77	0.85
20	41.7 ±1.7	27.1 ±2.8	56.2 ±5.7	42 ±5	0.56	0.75	1.06
21	172 ±7	179 ±12	84 ±15	64 ±12	0.67	0.77	0.90
22	1476 ±26	602 ±27	186 ±29	206 ±34	1.40	1.11	0.62
23	23 950 ±640	7680 ±380	1410 ±110	3250 ±300	0.90	2.31	2.01
24	2137 ±83	1028 ±67	675 ±83	614 ±82	0.55	0.91	1.30
25	4590 ±120	1173 ±60	860 ±81	808 ±87	0.75	0.94	0.99
26	429 ±17	123 ±9	258 ±22	201 ±20	0.50	0.78	1.22
27	2631 ±85	888 ±50	312 ±38	283 ±38	0.68	0.91	1.05
28	7010 ±190	2150 ±110	661 ±55	978 ±95	0.72	1.48	1.61
29	9610 ±280	3280 ±170	880 ±83	1360 ±150	0.66	1.55	1.86
30	1178 ±41	432 ±26	176 ±22	168 ±23	0.56	0.96	1.36
31	3660 ±100	39.8 ±9.1	2900 ±150	2690 ±190	0.89	0.93	0.82
32	62 200 ±2800	10 590 ±750	1640 ±230	6460 ±950	0.45	3.95	6.96
33	3720 ±120	1520 ±80	886 ±48	770 ±57	0.73	0.87	0.93
34	716 ±17	495 ±23	185 ±21	148 ±19	0.90	0.80	0.70
35	583 ±21	384 ±24	227 ±21	199 ±22	0.62	0.88	1.11
36	35 770 ±820	7280 ±330	2013 ±94	11 720 ±810	0.85	5.82	5.36
37	2044 ±58	377 ±21	562 ±36	764 ±63	0.85	1.36	1.27
38	755 ±28	222 ±15	612 ±39	501 ±41	0.60	0.82	1.07
39	1779 ±63	1112 ±69	912 ±85	784 ±85	0.64	0.86	1.06
40	597 ±20	205 ±14	410 ±37	364 ±37	0.79	0.88	0.87
41	8030 ±270	2893 ±170	720 ±160	1040 ±240	0.65	1.45	1.77
42	1208 ±46	377 ±24	215 ±21	283 ±32	0.54	1.32	1.92
43	956 ±31	227 ±15	375 ±40	363 ±43	0.74	0.97	1.03
44	939 ±25	382 ±21	431 ±42	387 ±43	1.09	0.90	0.65
45	368 ± 10	152 ±8	216 ±14	198 ±17	1.10	0.92	0.66
46	58.4 ±1.5	16.7 ±0.8	56.7 ±2.9	48.8 ±4.1	0.80	0.86	0.85
47	1921 ±72	840 ±55	829 ±90	688 ±83	0.58	0.83	1.13
48	6060 ±260	2840 ±200	2220 ±180	1820 ±180	0.49	0.82	1.31
49	3246 ± 100	695 ±40	516 ±62	1000 ±130	0.74	1.93	2.06
50	213 ±6	144 ±7	75.5 ±8.3	84 ±11	1.30	1.12	0.68
51	8390 ±250	4770 ±250	927 ±69	1056 ±95	0.79	1.14	1.13
52	25 460 ±590	15000 ±700	1048 ±50	3370 ±240	1.18	3.22	2.14
53	44 000 ±1100	27 300 ±1300	3540 ±370	8100 ±940	1.09	2.29	1.66
54	6800 ±300	3170 ±220	2720 ±200	2250 ±200	0.50	0.83	1.31
55	494 ±27	202 ±14	489 ±33	386 ±33	0.50	0.79	1.25
56	6510 ± 160	2580 ± 130	131 ± 30	317 ± 75	1.14	2.42	1.67
57	29 000 ± 660	3490 ± 160	1190 ± 54	5560 ± 380	1.23	4.67	2.99
58	48 100 ± 110	9080 ± 420	498 ± 59	3120 ± 400	1.26	6.26	3.90
59	9760 ± 310	780 ± 50	3320 ± 190	3750 ± 290	0.70	1.13	1.27
60	54 200 ± 1300	24 300 ± 1200	1593 ± 93	7440 ± 580	1.23	4.67	2.99

Calculated on the basis of the transmission experiment results, the values of correction factors fall within the range 0.51–6.96 cm²/g (values of coefficient *C* were normalized to the unit density so that they could be compared with all of the test samples). The values of correction factor lower than 1 indicates that the phenomenon of self-absorption in the sample takes place with a lesser intensity in comparison to the measurement standard used. In the majority of cases (ca. 85%) the correction factor ranged from 0.5–1.5 cm²/g, in other cases its value considerably exceeded 1.0. Due to the result of the radiation assessment risk, the determined correction is of particular importance for samples containing high concentrations of lead. Taking it into account may result in a change of radiation hazard category of a given place of work.

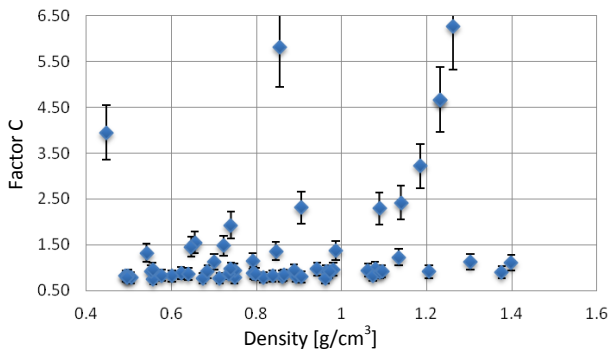


Figure 4. The dependence of factor *C* (non-standard) on the density of the sample

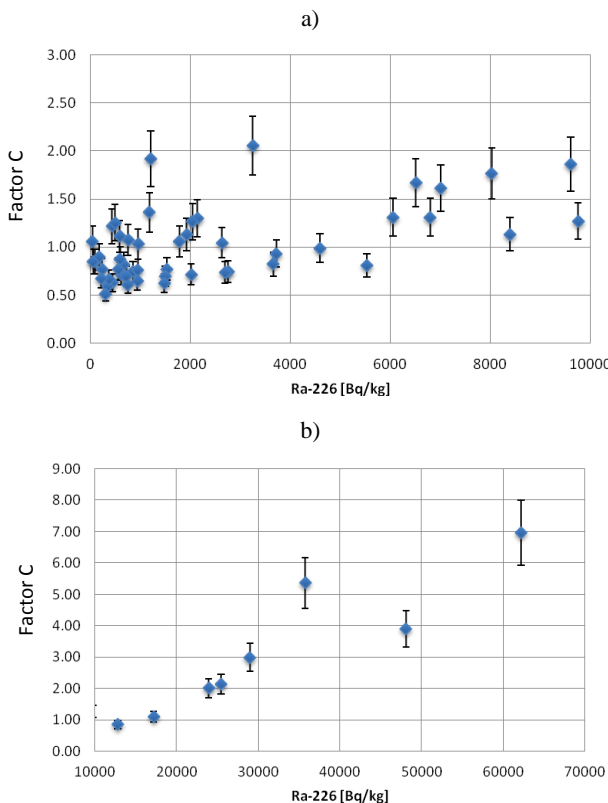


Figure 5 The dependence of factor *C* with a concentration of ²²⁶Ra: a – within the range 10 kBq/kg, b – within the range of 10 to 70 kBq/kg

Figure 4 shows the relationship between the value of the correction factor and the density of the sample. It is difficult to find an unequivocal relationship between these values

which means that the size of factor *C* is not directly dependent on density, or at least not only on it. Also, a relationship between the value of the correction factor and the concentration of the radioactive nuclides in the samples was searched for. In Figure 5a–b, a relation between the concentration ²²⁶Ra and the value of factor *C* is presented. For an improved clarity of the Figure, the data were divided into two groups: 0–10,000 and more than 10,000 Bq/kg ²²⁶Ra.

The presented graphs (Fig. 5a–b) show that, together with the increase of the concentration of ²²⁶Ra the value of correction factor *C* also increases. It can be explained by the mechanism of the formation of underground water sediments. Namely, the majority of sediments are formed as a result of co-precipitation of radium and barium in a form of barium-radium sulphate. Therefore, there should be a correlation between the concentration ²²⁶Ra and the content of barium (Ba). In the same process other metals can also be precipitated, which in turn influences the intensification of self-absorption phenomenon in the material of the sample.

5. CONCLUSIONS

²¹⁰Pb and its progeny are emitters of three types of ionizing radiation: α , β as well as γ . The mutual ratio of half-value periods of ²¹⁰Pb, ²¹⁰Bi and ²¹⁰Po, form a subseries of the radioactive series of ²³⁸U, causes these nuclides to reach the state of secular radioactive equilibrium relatively quickly. The physical and geochemical properties of lead mean that in the conditions, in which there is a possibility of matter being exchanged with the surroundings, its concentrations in various mediums does not necessarily depend upon the concentration of parent isotopes. ²¹⁰Pb is therefore an independent component among the factors of radiation risk. The need to closely monitor its concentration in terms of sediments formed from underground waters becomes justified.

60 samples of underground water sediments were examined in terms of a self-absorption phenomenon of energy 46.5 keV (²¹⁰Pb). On the basis of the measurements that were carried out, the following conclusions were formulated:

- a self-absorption phenomenon in the examined samples of sediments is important and considerably affects the result of lead ²¹⁰Pb determination in these samples
- calculated correction *C* oscillate within 0.51–6.96 cm²/g
- based on the relationship between the value of factor *C* and the concentration ²²⁶Ra in the sample, it can be concluded that a significant influence on the value of the self-absorption in the sample has the presence of barium and probably other metals

A large range of correlation factor *C* indicates the need to take into account its use in routine measurements¹. This is also supported by the fact that the most important (in terms of radiation protection) sediments, characterised by high concentrations of radioactive nuclides, are also characterised unit values significantly greater than the correction factor *C*. For these sediments there is a higher risk of making a mistake in

¹ In 2012, the measurement of the concentration of ²¹⁰Pb with the spectrometry of gamma radiation, taking into account the correction determined using the transmission method, was included in the scope of accreditation of the Silesian Centre for Environmental Radioactivity (Research and Calibration Laboratories of the Central Mining Institute, GIG).

the determination of concentration ^{210}Pb . After introducing changes into the control system of radiation hazard for miners, based on taking into account the ^{210}Pb concentration in mine sediments, a lack of the analysis of self-absorption phenomenon in the sample may lead to significant mistakes in its assessment.

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