

Regular Article

# Applicability of Oil Industry Waste Product in Building Industry from Radiological Point of View

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The oil industry currently produces huge amounts of drilling mud, and the re-use of these by-products are studied worldwide. In this study, the reusability of deposited drilling mud as a building material was investigated from a radiological point of view.

Terrestrial radionuclide concentrations were determined by gamma spectrometry, while the radon exhalations were evaluated by the accumulation method. The caused radiation doses were estimated by different calculation processes in cases in which the by-product was used as a building material.

The radionuclide contents were 31 (18-40) Bq/kg of <sup>226</sup>Ra, 35 (33-39) Bq/kg of <sup>232</sup>Th and 502 (356-673) Bq/kg of <sup>40</sup>K. The radon exhalation was 12 (6-17) mBq/kg, and the emanation coefficient of the samples was 6 (5-12) %.

According to the applied dose calculation models (indices), all of the results are below the limits; therefore, the use of drilling mud in the building industry exhibits no increased radiological risk. When comparing the applied indices with each other, it must be taken into consideration that the units indicate different dose values. In addition, their calculation is not consistent: excess dose rate vs. total dose Gy vs. Sv. etc.

**Key words:** gamma spectrometry, radiation dose, building material, drilling mud, radon exhalation

## 1. Introduction

The Earth's crust always contains radionuclides of natural origin in some amount. The main contribution to the natural terrestrial outdoor external exposure is the gamma ray emitting elements of the <sup>40</sup>K, <sup>238</sup>U, and the <sup>232</sup>Th decay chain. In world averages, the soil contains 33, 45 and 412 Bq/kg of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K, respectively<sup>1</sup>. Materials that include naturally occurring radionuclides,

such as <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K, are referred to as naturally occurring radioactive materials (NORM)<sup>1, 2</sup>. For each radionuclide, there is a NORM limit (10<sup>3</sup> Bq/kg for <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K) such that no regulation is required below this limit<sup>3-4</sup>. However, the effective dose limits should also be considered. The average soil and raw materials exhibit considerably lower nuclide concentrations than the mentioned NORM limits; in extreme cases, however, the nuclide content of the soil might exceed these limits. For example, Ghiassi-nejad *et al.*<sup>5</sup> reported a 4.2 × 10<sup>5</sup> Bq/kg <sup>226</sup>Ra concentration in the soil, and a 10<sup>3</sup> nGy/h absorbed gamma dose rate, while Vasconcelos *et al.*<sup>7</sup> reported 5.7 × 10<sup>5</sup> Bq/kg <sup>232</sup>Th in the soil. During anthropogenic industrial activities, the terrestrial nuclides may be

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enriched such that they approach the NORM limits, or even exceed them<sup>3, 7</sup>. The human-built environment surrounding mankind is created mainly from materials mined from the soil (concrete, brick, glass, metal); therefore, building materials contain the radionuclides present in the soil. Gamma radiation is produced by gamma ray emitting radionuclides of the soil, and the built environment causes different dose values when absorbed in humans<sup>8, 9</sup>. The worldwide average annual effective dose from external exposure due to terrestrial natural sources of radiation is 0.48 mSv, with 0.41 mSv stemming from exposure indoors (assuming an occupancy factor of 80%) and 0.07 mSv from exposure outdoors<sup>10</sup>. As lifestyle habits, including residence in buildings, may differ, this occupancy factor of 80% can be considered as conservative. Attention must be paid to the examination of building materials because humans spend the major part of their lives in buildings, and because the nuclide content of the building materials can be more easily regulated as compared to that of soil. Similarly, food stuffs<sup>11, 12</sup>, air<sup>13-15</sup> and drinking water<sup>16</sup> always contain radionuclides, which, when entering the human organism, causes internal radiation exposure. A major portion of internal radiation exposure, and nearly half of the total natural radiation exposure, is caused by radon and its daughter nuclides<sup>1</sup>.

<sup>222</sup>Rn is produced by the decay of radium. The greater part of the decay energy (4,871 MeV) is carried by the alpha particle, and a lesser part of it by the radon atom that is pushed back (86 keV). This energy is enough for a part of the radon to be released from the crystal lattice and to enter the pore space, and from there to move outside the material by the means of diffusion. Thus, the quantity of radon released from the material (called exhalation) is primarily determined by the <sup>226</sup>Ra concentration, the emanation and the diffusion inside the material. Physical parameters of the material that have an effect on the exhalation are as follows: moisture content, atmospheric pressure, radium distribution in grains, radiation damage, grain size and shape, temperature, external pore size, internal pore size and solid density (crystal structure and elements)<sup>17, 18</sup>. Radon mainly arrives in buildings from the soil below the building and from the building material. Consequently, a significant part of the internal radiation that human beings are exposed to originates from <sup>222</sup>Rn exhaled from the soil and the building material, and a lesser part originates from <sup>220</sup>Rn<sup>14</sup>.

The terrestrial gamma radiation and the internal radiation which humans are exposed to always have an effect, to some extent, on the human organism and the environment in which humans live. The negative physiological effect of average or low-level radiation exposure can only be examined with difficulty, since the

control group without this background dose can be represented with difficulty, or not at all. The evaluation is made even more difficult by the fact that there is no specific alteration that is a specific effect of radiation; hence, the harmful effect of radiation cannot be proved even statistically, or only above a certain value. There are several models used to estimate the health effects of radiation, including the LNT model (linear no-threshold model), which estimates the risk of low doses by means of extrapolation from the risk assessed at high doses, and hormesis, which attributes positive physiological effects to low doses.<sup>19</sup> Examination of the negative physiological effects of radiation in the case of doses that exceed the background value has been carried out several times, in cases related to radiation-based therapeutic treatments, nuclear disasters, uranium mines, etc.<sup>20, 21</sup>. Therefore, it is practical to compare the examined radionuclide concentrations with the world's average or the natural background, particularly when evaluating the health risk of the established dose spaces. Also, the EU BSS (European Basic Safety Standards)<sup>38</sup> and the IAEA (International Atomic Energy Agency)<sup>56</sup> take the existing exposure situation as local specialities into consideration, according to a higher exposure established due to the higher concentrations of the terrestrial (non-anthropogenic) nuclides.

In this study, the applicability in the building industry of drilling mud, the main component of which is bentonite, and which is stored in depositories as a side product, was investigated from a radiological aspect. Deposition is only one solution for drilling mud, which cannot be used for drilling any longer. Deposition can be achieved in a number of ways, including pits, mud boxes, land shaping, scattering over ground, discharging to the ocean, slurry injection, disposal in salt caverns, underground injection and range land disposal, which includes on-site burial (reserve pits). Deposition can be used in the recultivation of underground mines, in operating mines for backfilling, or insulation in abandoned workings. It can be used in finished mines for the safe storage of mining waste, or for quenching thermally active landfills<sup>22</sup>. Another possibility is in a land application in agriculture<sup>23</sup>. Before or during the agricultural application, the properties of the drilling mud can be improved by a bioremediation procedure<sup>24</sup>. Bentonite is used in the building industry for the production of watertight concrete and insulating concrete. The absorption ability, the ion-exchange capacity and the swelling behaviour of bentonite are important. Its use is considerable in building barrages and waste depositories; in such cases, the dose contribution for the population is negligible, however<sup>22</sup>. Harmless drilling sludge can be used in the production of building materials, including bricks, clay, building products, road surfaces, and drill

cuttings can be used in engineering work, by ignition calcination or by the addition of bricks and other building materials<sup>22, 25</sup>. With the use of bentonite, the application of cement can be reduced when concrete is produced<sup>26</sup>. A promising field is the phase change material application for low-temperature passive solar heating and cooling applications in buildings<sup>27</sup>. Due to economic considerations, a desire to reduce the ecological footprint, and a reduction of the number and size of the depositories, there is an ever increasing attention paid to the utilization and reuse of waste materials<sup>28-30</sup>. Furthermore, the demand for large quantities of inexpensive raw materials for cost reduction and environmental sustainability purposes has led to the recycling of industrial wastes, thus increasing concerns over public health. The classic example is phosphogypsum, which is a by-product generated from the large-scale production of phosphoric acid and is widely used instead of natural gypsum, e.g., in plasters, cements and reinforced glasses<sup>31</sup>.

## 2. Materials and methods

### 2.1. Sample collection

Samples were collected from the drilling mud depository of the MOL Group in Zalatárnok, Hungary (GPS coordinates: 46.682781, 16.729705). There are three depositories in the area. Two of them (90 × 150 m) had been already covered before the study, and so the third one (depository 70 × 30 m) was investigated. The deposited drilling mud is approximately 30,000 m<sup>3</sup>. Drilling mud samples were taken from four points on the midline of the depository (with 15 m distance from each other), and at two different depths (0.2m sample ID 1, 0.4m sample ID 2) to yield a 10 kg sample from each point. The examined depository had been opened five years before the sampling, and had been continuously filled for three years. The drilling mud samples were carried to the laboratory in plastic vessels.

### 2.2. Gamma spectrometry

The collected samples were dried to constant mass in an oven at a temperature of 105 °C. Before being stored for 30 days in airtight aluminium Marinelli vessels of 0.6 litre volume to reach secular equilibrium conditions between <sup>226</sup>Ra and <sup>222</sup>Rn (and their short-living decay products), the samples were crushed and ground in a mortar under a mesh of 0.63 mm diameter, and homogenized. The measurements were carried out using a GMX40-76 (ORTEC) HPGe device, while the spectra were evaluated using Maestro software. The resolution of the system was 1.85 keV at 1,332.5 keV and the samples were measured for 80,000 seconds. For the calibration, IAEA-326, the relevant reference material was used. The <sup>40</sup>K was measured directly (1,460 keV), while the <sup>232</sup>Th was

calculated from <sup>228</sup>Ac (338, and 911 keV) and <sup>208</sup>Tl (583keV), and <sup>226</sup>Ra was calculated from <sup>214</sup>Pb (352 keV) and <sup>214</sup>Bi (609 keV). For the applied measurement geometry, the minimum detectable activity (MDA) was 2, 2.4 and 20 Bq/kg for <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K respectively<sup>32</sup>.

### 2.3. Radon exhalation and emanation coefficient

The following section outlines the process of determination of the exhalation (Exh) and emanation coefficient (EC) of the collected drilling mud samples. The samples were ground to 630 nm and dried at 105 °C overnight, and the exhalation and emanation coefficient were determined using the method presented by Jonas *et al.*<sup>15</sup>. The applied system contained an accumulation chamber, air filter and radon-proof pump. Before measurements were taken, the chambers were purged with radon-free N<sub>2</sub> gas prior to the accumulation, in order to reduce the initial radon concentration (C<sub>0</sub>) to zero. The disturbing effect of toron was eliminated by a radon-proof tube of 3 m length and 4 mm internal diameter, placed in front of the AlphaGUARD, at a low 0.03 L/min flowrate. The radon activity concentration in the chambers was measured using an AlphaGUARD radon monitor (Saphymo) under closed loop circulation. The massic radon exhalation (Exh) and emanation coefficient (EC) were calculated using the following formulae<sup>33</sup>:

$$\text{Exh} = \frac{C \times V \times \lambda}{m \times (1 - \exp(-\lambda \times t))} \quad (1)$$

$$\text{EC} = \frac{V \times \text{Ceq}_{\text{Rn-222}}}{m \times \text{C}_{\text{Ra-226}}} \quad (2)$$

where

C: measured radon concentration C<sub>Rn-222</sub> [Bq/m<sup>3</sup>]

V: total volume of the measuring system [m<sup>3</sup>]

λ : <sup>222</sup>Rn decay constant [1/s]

t: accumulation time [s]

m: mass of the sample [kg]

Ceq<sub>Rn-222</sub>: radon concentration measured after secular equilibrium is reached [Bq/m<sup>3</sup>]

C<sub>Ra-226</sub>: radium activity concentration of the sample [Bq/kg]

MDA: 0.0015 mBq/kg-s, 5.4 mBq/kg-h

### 2.4. Indices used for evaluation

The calculation of indices used in the construction industry provides the answer to the question of whether it is possible to use drilling mud as a construction material or a raw material. Different indices were also compared to each other. Elimination of the radioactive components of the drilling mud is too difficult or too expensive. Therefore, nuclide contents may be a primary limiting factor of utilization and valorization of drilling mud<sup>34-38</sup>.

### Radium equivalent concentration $Ra_{eq}$

For the comparison of materials containing  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  the  $Ra_{eq}$  (Bq/kg) (radium equivalent concentration) is widely used.  $Ra_{eq}$  is calculated using the equation 3.

$$Ra_{eq} = C_{Ra-226} + 1.43 \times C_{Th-232} + 0.077 \times C_{K-40} \quad (3)$$

where  $C_{Ra-226}$ ,  $C_{Th-232}$  and  $C_{K-40}$  are the activities concentration (Bq/kg) of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ , respectively<sup>39, 40</sup>.

### I index

For utilization in the construction industry, the composition of the finished product must meet the I index. I is calculated using Equation 4 below. Index I determines the dose caused by indoor external gamma radiation in addition to outdoor external radiation in the case of a building built using the examined building material. This model is conservative. Each wall, floor and ceiling is made from concrete, and the room dimensions are  $3 \times 4 \times 2.5$  m without windows and doors. For  $I \leq 1$  the annual dose is  $\leq 1$  mSv; for  $I < 0.5$ , the annual dose is  $< 0.3$  mSv<sup>38, 42, 43</sup>.

$$I = \frac{C_{Ra-226}}{300} + \frac{C_{Th-232}}{200} + \frac{C_{K-40}}{3000} \quad (4)$$

where:  $C_{Ra-226}$ ,  $C_{Th-232}$ ,  $C_{K-40}$ ,  $Ra-226$ ,  $Th-232$  and  $K-40$  concentrations are in [Bq/kg]. The EU BSS nominates and utilizes this index.

### H indices

The H indices are suitable for the calculation of the external and internal radiation dose originating from the building material on those residing in the building. These are defined by Equations 5 and 6. In the case that  $H_{ex} \leq 1$ , the annual extra dose resulting from external gamma radiation caused by the building is  $\leq 1.5$  mSv/year. If  $H_{in} \leq 1$ , the internal dose due to radon and its progenies is  $\leq 1.5$  mSv/year<sup>39, 43-45</sup>

$$H_{ex} = \frac{C_{Ra-226}}{370} + \frac{C_{Th-232}}{259} + \frac{C_{K-40}}{4810} \quad (5)$$

$$H_{in} = \frac{C_{Ra-226}}{185} + \frac{C_{Th-232}}{259} + \frac{C_{K-40}}{4810} \quad (6)$$

### Representative Level Index (RLI)

The  $\gamma$ -ray representative index is used as a tool to estimate the radiation hazards in building materials associated with natural radionuclides. It is defined by Equation 7. An RLI  $\leq 1$  in the case of flats, i.e. block apartments, indicates 0.1 mSv, and 0.03 mSv in the case of single-family houses due to the extra dose from the building material<sup>46</sup>.

$$RLI = \frac{C_{Ra-226}}{150} + \frac{C_{Th-232}}{100} + \frac{C_{K-40}}{1500} \quad (7)$$

### Activity Utilization Index (AUI)

To facilitate the calculation of dose rates in the air from different combinations of the three radionuclides in building materials samples, and by applying the appropriate conversion factors, an activity utilization index (AUI) can be constructed, as given by Equation 8:

$$AUI = \frac{f_U \times C_{Ra-226}}{50} + \frac{f_{Th} \times C_{Th-232}}{50} + \frac{f_K \times C_{K-40}}{500} \quad (8)$$

The values  $f_U$  (0.462),  $f_{Th}$  (0.604) and  $f_K$  (0.041) are the fractional contributions to the total dose rate in the air due to gamma radiation from the actual concentrations of these radionuclides. The recommended safe limit is an index value of 2, which corresponds to an annual effective dose of  $< 0.3$  mSv/y<sup>47</sup>.

### Outdoor absorbed gamma dose rate ( $D_{out}$ , [nGy/h])

If drilling mud is used for filling or outdoor cover from the radionuclide contents of the upper 20 cm of the soil, the absorbed gamma dose rate at 1 m can be calculated using Equation 9<sup>8</sup>.

$$D_{out} = 0.4551 \times C_{Ra-226} + 0.5835 \times C_{Th-232} + 0.0429 \times C_{K-40} \quad (9)$$

at 1 m above the ground surface is assessed from the  $\gamma$ -radiation originating from  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ , assumed to be equally distributed in the ground.

### Indoor gamma dose rates ( $D_{in}$ , [nGy/h])

The indoor absorbed gamma dose rate calculated from the nuclide contents of the building is given by the Equation 10<sup>48</sup>.

$$D_{in} = 0.92 \times C_{Ra-226} + 1.1 \times C_{Th-232} + 0.081 \times C_{K-40} \quad (10)$$

where the population-weighted average is 84 nGy/h<sup>48</sup>.

### Excess absorbed gamma dose ( $D_{ex}$ [nSv/h], $E_{ex}$ [mSv/y])

In the case of assumed residence in the buildings, we calculated how the excess absorbed gamma dose caused by terrestrial radionuclides in the building material would affect a representative person, according to the IAEA safety guide<sup>10</sup>. The model supposed that the building was made of concrete with a density of 2,300 kg/m<sup>3</sup>. The floor was 0.3 m thick and the walls were 0.2 m thick. For each of the indices, it can be said that in the case of one unit of activity, the contribution of  $^{40}\text{K}$  to the sum

**Table 1.** Drilling mud Activity concentration, Radon exhalation, Radon emanation coefficient

Sample ID Drilling mud	Activity concentration [Bq/kg]			Exh [mBq/kg]	EC [%]
	<sup>226</sup> Ra	<sup>232</sup> Th	<sup>40</sup> K		
A/1/1	39 ± 2	35 ± 4	582 ± 6	13 ± 2	5 ± 1
A/1/2	40 ± 3	37 ± 4	632 ± 7	15 ± 2	5 ± 1
A/2/1	19 ± 2	34 ± 3	392 ± 5	16 ± 2	12 ± 2
A/2/2	36 ± 2	35 ± 4	673 ± 7	17 ± 2	7 ± 1
A/3/1	33 ± 2	34 ± 4	417 ± 5	10 ± 2	4 ± 1
A/3/2	37 ± 2	36 ± 4	595 ± 5	10 ± 2	4 ± 1
A/4/1	24 ± 2	37 ± 4	356 ± 4	6 ± 1	4 ± 1
A/4/2	18 ± 2	35 ± 4	368 ± 4	8 ± 1	7 ± 1
<b>average</b>	<b>31 (18-40)</b>	<b>35(33-39)</b>	<b>502 (356-673)</b>	<b>12 (6-17)</b>	<b>6 (5-12)</b>
SD	9	1	131	4	3
median	35	35	500	12	5
World soil average <sup>1)</sup>	32	45	412		5-70
Hungary soil average <sup>48)</sup>	33	28	370		
NORM	10 <sup>3</sup>	10 <sup>3</sup>	10 <sup>3</sup>		

dose is lower by one order of magnitude as compared to that of <sup>226</sup>Ra or <sup>232</sup>Th. The reason for this is that in the decay chain of <sup>226</sup>Ra and <sup>232</sup>Th, several radiating nuclides are formed, while <sup>40</sup>K decays to stable Ar and Ca.

### Excess Lifetime Cancer Risk (ELCR).

Excess lifetime cancer risk (ELCR) is calculated using Equation 11:

$$ELCR = AEDE \times DL \times RF \quad (11)$$

where AEDE is the annual effective dose equivalent, DL is a duration of life (70 years), and RF is the risk factor (1/Sv), or the fatal cancer risk per sievert. For stochastic effects, ICRP 60 uses the value of 0.06 for the public<sup>9,50)</sup>.

## 3. Results and discussion

### 3.1. Measured activity concentrations

The activity concentrations of the drilling mud are shown in Table 1. The values were 31 (18-40) Bq/kg for <sup>226</sup>Ra, 35 (33-39) Bq/kg for <sup>232</sup>Th and 502 (356-673) Bq/kg for <sup>40</sup>K, while the average calculated Ra<sub>eq</sub> was 121 (96-141) Bq/kg. The magnitude of each radionuclide concentration meets either the world soil average or the average value of soil for Hungary<sup>1, 48)</sup>. Based on the IAEA's world average for building materials<sup>10)</sup> 40, 30 and 400 Bq/kg, and according to the results of Szabó *et al.* in Hungary, the artificial building material (brick, concrete, gas silicate) was 202, 29 and 204 Bq/kg for <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K, respectively. In our study, the <sup>40</sup>K value of the drilling mud was slightly above the world soil average of 412 Bq/kg (same range as the average for the typically used building material<sup>10, 46)</sup>), but its contribution to the absorbed gamma dose rate value is one order of magnitude lower than that of <sup>226</sup>Ra

or <sup>232</sup>Th (see Equations 9 and 10). The values measured were three magnitudes lower than the regulatory limit for the NORM category<sup>3)</sup>. Samples taken from point A1 that had the highest <sup>226</sup>Ra concentration also had nuclide concentrations similar to the environmental samples, and had <sup>226</sup>Ra content lower than that of artificial building materials of previous measurements<sup>42, 44, 45, 47)</sup>. Previous measurements confirmed that the drilling mud and scale have significantly lower activity concentrations than sludge<sup>51)</sup>.

The average value of exhalation is 12 (6-17) mBq/kg which is normal. Exhalation values below 10 mBq/kg were reported for manganese clay samples baked at 750 °C, or red mud samples baked at 1,200 °C<sup>52)</sup>. Elzain *et al.*<sup>53)</sup> measured the exhalation of 12 different raw materials and finished products used for building materials to be 2.8-11.25 mBq/kg. However, the exhalation value of normal building materials can reach even 31–271 mBq/kg. The emanation coefficient of the examined drilling mud samples was 6 (4-12)%. Compared to earlier examinations, this value can be considered low, as the emanation of manganese clay baked at 650 °C was 16 %<sup>52)</sup>, that of Hungarian clay samples was 8–34%<sup>18)</sup>, waste material from the oil industry (scale, mud, etc.) was 47 (15-57)%<sup>36)</sup> and typical emanation coefficients for rocks and soils ranged from 5 to 70%<sup>48)</sup>. The low EC in the drilling mud was probably caused by its cleaning treatment, during which the <sup>226</sup>Ra was removed and washed from the surface of the granules; furthermore, the drilling mud has a high level of silicate content and low organic matter contents. This is in accord with the findings of Greeman *et al.*<sup>54)</sup>, in which the <sup>226</sup>Ra present in the coating of the granules was shown to have the highest EC value. Furthermore, a higher EC value was measured in the exchangeable cations and organic matter

**Table 2.** Calculated indices

	Sample	I index	H <sub>ex</sub>	H <sub>in</sub>	RLI	AUI	D <sub>in</sub>	D <sub>out</sub>	E <sub>out</sub>	E <sub>in</sub>	ELCR		adult		children		infants	
							nGy/h	nGy/h	mSv/y	mSv/y	×10 <sup>5</sup>	×10 <sup>4</sup>	D <sub>ex</sub>	E <sub>ex</sub>	D <sub>ex</sub>	E <sub>ex</sub>	D <sub>ex</sub>	E <sub>ex</sub>
Drilling mud	A/1/1	0.5	0.4	0.5	0.8	0.8	122	63	0.1	0.7	33	25	45	0.3	52	0.4	58	0.4
	A/1/2	0.5	0.4	0.5	0.9	0.9	129	67	0.1	0.7	34	27	50	0.4	58	0.4	65	0.5
	A/2/1	0.4	0.3	0.3	0.6	0.6	87	45	0.1	0.5	23	18	20	0.1	23	0.2	26	0.2
	A/2/2	0.5	0.4	0.5	0.9	0.8	126	66	0.1	0.7	34	26	48	0.3	55	0.4	62	0.4
	A/3/1	0.4	0.3	0.4	0.7	0.7	102	53	0.1	0.6	27	21	31	0.2	35	0.3	40	0.3
	A/3/2	0.5	0.4	0.5	0.8	0.8	122	63	0.1	0.7	33	25	45	0.3	52	0.4	58	0.4
	A/4/1	0.4	0.3	0.3	0.6	0.7	92	48	0.1	0.5	25	19	24	0.2	27	0.2	31	0.2
	A/4/2	0.4	0.3	0.3	0.5	0.6	85	44	0.1	0.5	23	17	19	0.1	22	0.2	24	0.2
	average	0.4	0.3	0.4	0.7	0.8	108	56	0.1	0.6	29	22	35	0.3	40	0.3	45	0.3
	SD	0.1	0.1	0.1	0.1	0.1	19	10	0.0	0.1	5	4	13	0.1	15	0.1	17	0.1
median	0.5	0.3	0.4	0.7	0.8	111	58	0.1	0.6	30	26	38	0.3	43	0.3	49	0.3	
World soil av.	0.5	0.3	0.4	0.7	0.9	112	59	0.1	0.6	30	20	39	0.3	44	0.3	50	0.4	
Hungary soil av.	0.4	0.3	0.4	0.6	0.7	91	47	0.1	0.5	20	20	24	0.2	27	0.2	30	0.2	
NORM	9	7	10	12	21	2101	1082	1	12	560	430	1481	10	1692	12	1904	13	

than in the silt or clay fraction. Due to the low exhalation and emanation coefficient, the drilling mud, when mixed with other materials, can reduce the radon exhalation of the finished product in the case of application in the building industry. While exhalation and emanation coefficient values are low, the applied measurements in this range are affected by significant error levels (10-17%). However, by conservative evaluation, the values increased by the errors are also low.

The calculated indices are summarized in Table 2.

### 3.2. Evaluation of the indices

#### I index

The I index is 0.4 (0.4-0.5), which is less than 1, and thus it meets with the current regulation. The annual dose from the building material is thus less than 1 mSv. In case of I < 0.5, the dose from the building material < 0.3 mSv/y. The I index of the drilling mud is similar to the average of the natural building materials from Hungary (0.35); however, this is only half of the value (0.89) of the artificial building materials according to Szabó *et al.*<sup>45)</sup>. Based on a survey from the EU, where the I index of 1,953 samples from 24 countries were examined, 86% of the investigated bricks had 0.5 < I < 1, and 57% of the concrete had 0.5 < I < 1<sup>55)</sup>. Therefore, the building materials made from our drilling mud (what was studied) have lower I index values than common building materials. Based on our results, drilling mud can be used in building materials without any restriction. The E<sub>in</sub> value indicates a higher annual dose 0.5 (0.4-0.6) mSv/y than that calculated from I index, 0.3 (0.2-0.5) mSv/y. Based on the IAEA safety guide, the E<sub>ex</sub> of infants 0.3 (0.2-0.5) mSv/y is the same as a calculated dose from the I index.

#### H indices

H<sub>in</sub> 0.4 (0.3-0.5) < 1, H<sub>ex</sub> 0.3 (0.3-0.4) < 1, which means that both internal and external radiation is less than 1.5 mSv/y.

#### Representative Level Index (RLI)

RLI = 0.7 (0.5-0.9) < 1, which means that the extra dose that is associated with the natural radionuclide in building materials is < 0.1 mSv/year in the case of a block apartment, and < 0.03 mSv/year in the case of single-family houses.

Therefore, RLI is the most strict index; however, none of the measured samples reach the 1 value. The other indices show a higher annual effective dose than does the RLI (0.1 and 0.03 mSv/y).

#### Activity Utilization Index (AUI)

AUI = 0.8 (0.6-0.9) < 2 means that the annual effective dose < 0.3 mSv/y. This value is the same as that calculated based on the IAEA safety guide and I index.

#### Indoor absorbed gamma dose rates (D<sub>in</sub>)

D<sub>in</sub> = 108 (85-129) nGy/y, which is slightly higher than the population-weighted world average of 84 nGy/h<sup>1)</sup>.

The value of the D<sub>in</sub> was two times higher than the value of the D<sub>ex</sub>. The D<sub>ex</sub> takes the shielding effect into consideration as well, and only gives the excess dose from the building over the value of the outdoor dose. In this D<sub>in</sub> level, the values were higher than in the other indices.

#### Outdoor absorbed gamma dose rate (D<sub>out</sub>)

D<sub>out</sub> = 56 (44-67) nGy/h. Note that the D<sub>out</sub> is unaffected by the value of the cosmic radiation, so it cannot be compared to the outdoor absorbed gamma dose rate. The outdoor terrestrial gamma dose rate (population-weighted average) equals 59 nGy/h<sup>48)</sup>.

#### Annual Effective Dose E<sub>in</sub>, E<sub>out</sub>

E<sub>in</sub> = 0.6 (0.5-0.6) mSv/y, which is calculated from D<sub>in</sub> and so the same conditions discussed above apply to it.

E<sub>out</sub> = 0.1 mSv/y,

$E_{\text{total}} = E_{\text{in}} + E_{\text{out}} = 0.7 \text{ mSv/y}$  is slightly higher than the 0.48 mSv/y world average<sup>(10)</sup>.  $D_{\text{out}}$  is unaffected by the values of the cosmic radiation. This value is negligible compared to the 2.4 mSv/y.

#### Excess Lifetime Cancer Risk (ELCR).

**Average**  $ELCR_{\text{out}} = 29 \times 10^5$ . This is the same as the value calculated from the word average of the radionuclide content of the soil  $30 \times 10^{5.1}$ .

**Average**  $ELCR_{\text{in}} = 22 \times 10^4$ . It can be seen that, compared to the  $ELCR_{\text{out}}$ , the risk is one order of magnitude higher. The cause of this is the 0.8 fraction of time spent indoors, and the radiation caused by the walls and ceiling.

#### Excess absorbed gamma dose rate ( $D_{\text{ex}}$ )

The calculated average  $D_{\text{ex}}$  is 38, 43 and 49 nSv/h for adults, children and infants, respectively.  $D_{\text{ex}}$  shows only the excess dose caused by the building material.

#### Annual effective excess dose ( $E_{\text{ex}}$ )

Average  $E_{\text{ex}}$  is 0.25, 0.28 and 0.32 mSv/y for adults, children and infants, respectively, which is lower than the indoor terrestrial natural sources of radiation exposure, 0.41 mSv<sup>(10)</sup>.

## 4. Conclusion

Drilling mud from Hungarian oil drilling—currently a waste product—was examined from a radiological perspective. Terrestrial nuclide concentrations were determined by gamma spectrometry. The radon exhalation of the samples was determined by the accumulation method. The radiological risk of building materials and other indices were estimated by calculation. The values were 31 (18-40) Bq/kg  $^{226}\text{Ra}$ , 35 (33-39) Bq/kg  $^{232}\text{Th}$ , 502 (356-673) Bq/kg  $^{40}\text{K}$ , while the average calculated  $^{226}\text{Ra}_{\text{eq}}$  was 121 (96-141) Bq/kg. The  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  content is slightly lower than the world average but practically identical with it; however,  $^{40}\text{K}$  content exceeds the world average to a small extent.  $^{40}\text{K}$  contributes to the established dose rates to a lesser extent, and thus no dose increase should be experienced. Our findings were that I index = 0.4 (0.4-0.5),  $H_{\text{ex}} = 0.3$  (0.3-0.4),  $H_{\text{in}} = 0.4$  (0.3-0.5), RLI = 0.7 (0.5-0.9), AUI = 0.8 (0.6-0.9),  $D_{\text{in}} = 108$  (85-129) nGy/y,  $D_{\text{out}} = 56$  (44-67) nGy/y,  $E_{\text{in}} = 0.6$  (0.5-0.7) mSv/y,  $E_{\text{out}} = 0.1$  mSv/y,  $ELCR_{\text{out}} = 29 \times 10^5$ ,  $ELCR_{\text{in}} = 22 \times 10^4$  and that  $D_{\text{ex}} = 38, 43$  and 49 nSv/h and  $E_{\text{ex}} = 0.25, 0.28$  and 0.32 mSv/y for adults, children and infants, respectively. The radon exhalation was 12 (6-17) mBq/kg, and the emanation coefficient of the samples was 6 (5-12) %. It can be established that the use of drilling mud in the construction industry exhibits no increased radiological risk. There are several indices to describe the radiological risk of the building materials, but note that these indices are not compatible with each other just by means of a simple conversion. Their calculation is not consistent; one

refers only to the excess dose, while the other represents the total dose. In addition, one refers the dose in Gy the other in Sv units. In the case in which I index = 1, the dose rate caused by the building material is 1 mSv/y, whereas for AUI < 2, it is < 0.3 mSv/y. Thus, during radiological risk evaluation the value of the dose should be taken into account, not the value of the index.

## Conflict of Interest Disclosure

The authors declare that they have no conflict of interest.

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